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LIQUEFACTION OF NOVA SCOTIA COALS
PHASE VI

STUDIES ON INTEGRATED TWO-STAGE THERMAL/CATALYTIC LIQUEFACTION

FINAL REPORT

by

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SUMMARY

The sixth phase of a research program on the direct liquefaction of Nova Scotia coal has been completed. This phase of the work was concentrated on integrated two-stage thermal/catalytic liquefaction. Process studies were initiated using unimodal micropore catalysts in the second-stage reactor.

During Phase VI, our bench scale coal liquefaction unit was modified and expanded for two-stage operations. A total of twenty-five liquefaction experiments were conducted with Lingan Harbour seam coal to investigate the effects of operating conditions on coal conversion and raw product distribution. Experiments were conducted in a once-through mode using hydrogenated anthracene oil, 9% H, as reaction solvent. Two commercial hydroprocessing catalysts, Katalco 504-K nickel-molybdenum and Katalco 550 nickel-tungsten have been tested. Experimental results have shown that coal space velocity, total pressure and catalytic reactor temperature significantly affect product composition and hydrogen consumption. Katalco 504-K exhibited a slightly higher degree of activity for hydrogenation, denitrogenation and deoxygenation reactions. Recent work on separator bottoms product suggests that Katalco 550 has superior cracking ability.

Two-stage thermal/catalytic operations with Prince Hub seam coal produced higher C_4^+ liquid yield and lighter products than operations with Lingan Harbour seam coal. Maximum C_4^+ liquid yields from Prince and Lingan coals were 66 and 62 wt.%, respectively, based on maf coal plus hydrogen added. In the former case, 55 wt.% of the maf coal was converted to pentane soluble oil compared with 47 wt.% in the latter. The higher reactivity of Hub seam coal is primarily attributed to its lower rank.

The next phase of work will concentrate on process optimization, coal reactivity and catalyst evaluation studies. Additional test work is planned to complete our evaluation of unimodal catalysts and to assess the performance of at least one bimodal catalyst with Prince Hub, Lingan Harbour and Donkin-Morien Harbour seam coals. On completion of the thermal/catalytic studies, the first stage thermal dissolver will be converted to a catalytic dissolver. Following this, a test program will be undertaken to assess the potential of two-stage catalytic liquefaction applied to Sydney area coals.

INTRODUCTION

The Organization of Petroleum Exporting Countries was established in 1960 to gain control over oil prices from the multinational oil companies. By 1973, the economic well-being of industrial nations that were dependent wholly or in part on foreign sources of crude oil was threatened by energy crises, feedstock shortages and soaring fuel costs. The unpredictability of the international oil market influenced Western governments to intensify conservation efforts and expand their domestic energy base. As a consequence of the energy strategies formulated by Western nations, considerable attention has been focused on the production of liquid fuels from coal in recent years.

Historically, two routes have been used to produce hydrocarbon liquids from coal - a direct or hydroliquefaction route, and an indirect or synthesis route. During the past fifteen years, significant advances have been made relating to the production of synthetic liquid fuels via coal liquefaction. Within the past decade, large-scale pilot plants have been built in the United States and Germany to determine the economic feasibility of direct liquefaction processes, e.g., Exxon Donor Solvent, H-Coal and Ruhrkohle/Veba.

Since 1982, however, crude oil prices have stabilized and are actually decreasing substantially. This has resulted in the cancellation or slow-down of most large-scale synfuels projects in North America. Nevertheless, a modest level of research is being maintained to pursue

second generation processes. In other parts of the world, industrialized nations, including Germany and Japan, are still actively conducting programs to further develop coal liquefaction technology.

In Canada, our way of life is highly dependent on petroleum liquids. Since our existing crude supplies are not expected to meet our future liquid fuel demands, it seems that we will have to rely on synthetic liquid fuels to some degree in the next few decades. Thus, should it be economically attractive or essential, abundant coal reserves are available in Canada for conversion to synthetic liquid fuels. Because long lead times are involved in process evaluation, detailed design and plant construction, it is essential to understand the basic concepts of the promising liquefaction processes and verify their potential yield structures as they apply to Canadian coals.

Process studies relating to the direct liquefaction of Nova Scotia high-volatile bituminous coals were initiated at the Nova Scotia Research Foundation Corporation (NSRFC) in 1978. Initially, efforts were concentrated on a low-severity extractive coking process (1,2,3). From 1980 to 1984, experimental work was conducted on medium to high pressure donor solvent extraction (4,5) and single-stage catalytic hydrogenation (6). Yield data has been developed over a wide range of operating conditions using bench-scale batch and continuous-flow hydroprocessing units.

Test results have shown that single-stage catalytic hydrogenation produces higher oil yields than donor solvent extraction. Furthermore, it

has been shown that considerable control can be exercised over product distribution by varying process severity. Once-through experiments conducted with Lingan Harbour seam coal and commercial nickel-molybdenum hydrotreating catalyst over medium to high severity operating conditions produced high coal conversions, 90-92%, while oil yields ranged from 30-47%.

Direct catalytic coal liquefaction processes, such as H-Coal, are single-stage processes. The catalyst serves several functions and the severe operating conditions required for one function might not be needed for other functions. Recent process developments in the United States are centered on liquefying coal in two separate reaction zones. Two-stage processing offers the versatility of controlling coal dissolution and primary product upgrading under different operating conditions.

During the first half of 1985, our bench-scale coal liquefaction unit was modified and expanded for integrated two-stage operations. The closely coupled two reactor system was built because two-stage processing offers the potential of higher distillate yields and improved hydrogen utilization. Details pertaining to the system are presented in this report.

The two-stage unit was operated in a thermal/catalytic mode from June to November of 1985. A total of twenty-six experiments were conducted with commercially available nickel-molybdenum and nickel-tungsten hydroprocessing catalysts in the second stage reactor. Both catalysts have unimodal pore size distributions with pores of about 100 angstroms

diameter. The effects of major process variables on coal conversion and product distributions were investigated. The results of the tests are given herein.

TASK 1 - EQUIPMENT MAINTENANCE

Actual work on Phase VI began in October of 1984. The first task involved inspection and maintenance of existing systems. Efforts were concentrated on the hydrogen compression facility, the fixed-bed catalytic hydrotreater and the continuous coal liquefaction unit. These systems were operated frequently during the previous nine-month period, Phase V.

In the hydrogen compression and storage area, attention was centered on the Kemlon two-stage compressor. This unit is 3 horsepower with a design capacity of 28 L/min from 1.5 MPa to 28 MPa. Routine maintenance includes changing the gear oil periodically and adding actuating fluid as required. During this turnaround period, suction and discharge valves were removed and cleaned, O-ring seals were replaced and a new stainless steel diaphragm was installed in the second-stage head. In addition, minor adjustments were made to the injector and relief valves on the actuating fluid compensator system.

The fixed-bed catalytic hydrotreater has been described in an earlier report (3). On turnaround, the reactor was removed from the system, disassembled and charged with fresh catalyst. Heating tapes on the feed vessel and liquid transfer lines between the feed vessel and the reactor inlet were replaced. The high pressure liquid metering pump was given a general overhaul and new packing rings were installed in the pump body.

A complete description of the coal liquefaction unit as configured in Phase V is presented elsewhere (5,6). In preparation for

planned modifications, various sections of the single-stage unit were disassembled and inspected in November, 1984. Routine maintenance was carried out on the low pressure slurry circulation pump, high pressure let-down valves, block valves and automatic process control equipment.

Slurry transfer lines in the low pressure feed circuit and the high pressure liquefaction section were found to be essentially free of solids. However, small deposits of solids saturated with oil were found in the stirred tank reactor and the primary high pressure separator located downstream of the reactor. In both cases, the material was easily removed. The gummy deposits were washed with an excess of pentane to remove the oil and then dried.

Approximately 6 g of dry solids were recovered, 4 g from the reactor and 2 g from the separator. On further examination, solids from the reactor were found to contain 18.1% volatile matter, 22.9% fixed carbon and 59.0% ash. Solids from the separator contained 46.5% volatile matter, 9.7% fixed carbon and 43.8% ash. The inspection of high pressure lines and vessels was encouraging in that no signs of coke deposition were observed after several months of operation.

TASK 2 - DESIGN INTEGRATED TWO-STAGE COAL LIQUEFACTION UNIT

2.1 Background

The purpose of Task 2 was to specify the design changes and equipment required to expand our existing coal liquefaction unit for integrated two-stage thermal/catalytic operations. Fairly extensive modifications were required to the high pressure reaction zone and the low-pressure slurry feed section.

The main priority in the high pressure reactor section was the addition of a 3.8-liter stirred tank reactor (STR). The 3.8-liter STR was actually purchased during Phase V. Subsequently, it was modified to function as a catalytic reactor. This was accomplished by installing a catalyst basket assembly and an impeller/baffle system in the reactor. The capacity of the catalyst basket is about 600 cc. The components required were manufactured by Autoclave Engineers Inc. and are commonly known as Robinson-Mahoney style internals.

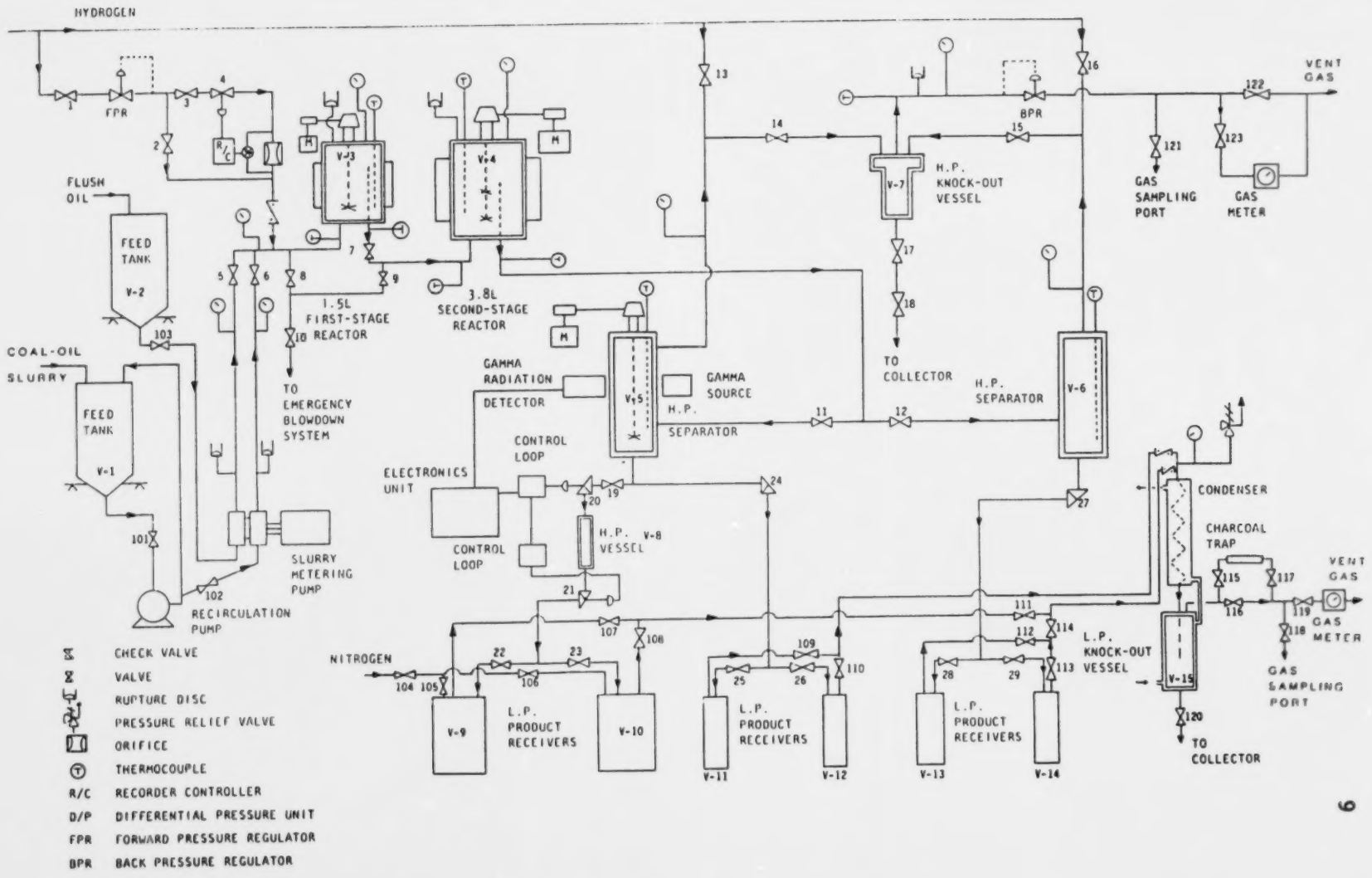
During operation, hot reacting slurry is constantly recirculated within the gradientless catalytic reactor. Slurry is forced through the annular basket and consequently across the catalyst by the impeller situated in the open area in the center of the basket. Slurry product and gases exit the reactor via an overflow pipe. This configuration permits catalyst to be used in bead or extrudate form. Furthermore, catalyst can be sulphided in-situ prior to coal liquefaction runs.

2.2 Description of Two-Stage Unit

The integrated two-stage coal liquefaction unit is designed for diverse applications. A schematic diagram of the system is presented in Figure 1. The design provides a closely coupled two-stage reaction zone and a separate feed circuit for flushing the unit. During Phase VI, the unit was operated with a thermal first stage and a catalytic second stage. The apparatus is suitable for high pressure (24 MPa) use and is capable of processing 0.7-3.0 kg per hour of coal/oil slurry.

In the system, coal and oil are mixed batchwise in a 20-liter feed vessel. The coal/oil slurry is continuously circulated by a rotary pump. This arrangement prevents settling of coal fines and provides suction pressure to the slurry metering pump. Hydrogen is added and the mixture is fed to the 1.5-liter first-stage reactor. The reactor is heated by a two-zone electric heater. Effluent from the first stage flows to the second stage reactor. Downstream of the reaction zone, products are passed to a hot separator. The overhead vapours and gases from the hot separator pass to a cold knock-out vessel. Light liquid products are condensed in the cold knock-out and a vent gas stream is provided through a back pressure regulator. An automated high pressure let-down system controls the rate at which slurry product is withdrawn from the bottom of the hot separator. The bottoms slurry is flashed at atmospheric pressure and collected. Vapours and gases from the low pressure product receivers are passed through a water-cooled knock-out vessel and condenser. Vent gas streams are monitored with gas meters.

Figure 1 - SCHEMATIC DIAGRAM OF TWO-STAGE COAL LIQUEFACTION BENCH UNIT



TASK 3 - PROCUREMENT OF NEW EQUIPMENT

Activities associated with this task included the selection and procurement of new equipment. Major pieces of equipment required for the two-stage liquefaction unit were as follows:

(1) Solid-state temperature controller for the second-stage reactor. The reactor complete with furnace was actually purchased during Phase V.

(2) Robinson-Mahoney style internals for the 3.8-liter STR mentioned above.

(3) Bolted closure pressure vessel, nominal capacity 7.6-liters, required as an emergency blow-down reservoir.

(4) Digital pressure indicator system for precise measurement of differential pressure across the reaction zone.

In addition, a new hydrogen compressor was acquired to replace our aging unit, and to provide increased capacity for longer term tests. The new unit is a single-stage diaphragm compressor. It was supplied by Pressure Products Industries (PPI). The design capacity of the 7.5 horsepower unit is 114 L/min from 3.5 MPa to 34.5 MPa.

TASK 4 - INSTALLATION OF NEW EQUIPMENT

According to the original work plan, assembly of the two-stage liquefaction unit was scheduled for completion by February 15, 1985. However, prolonged delivery times associated with key pieces of equipment forced the completion date for assembly to slip by about three months.

In order to provide adequate space for the closely coupled two reactor configuration, it was necessary to relocate the low pressure slurry feed system. Electrical and mechanical work associated with relocating the low pressure equipment began in January, 1985. Over a period of several weeks, the low pressure section was rebuilt to accommodate separate slurry and flush oil feed systems.

Additional steel framework required to support the reactor section and the Bran and Lubbe metering pump was fabricated and installed in February. Following this, the metering pump and the first-stage reactor were installed. Early in March, efforts were concentrated on reassembly of equipment in the high pressure separation and let-down sections of the unit. Modifications were made to the separator overheads section to simplify the collection of separator overhead liquids. By the end of March, the furnace and temperature controller for the 3.8-liter second-stage reactor were installed, and additional circuits were provided to control heat tracing on product transfer lines.

Thermocouples, rupture discs and vent lines were added to the rig during the first two weeks of April. The digital pressure indicator system and the 7.6-liter blow-down reservoir were installed by the end of

April. During the first two weeks of May, the 3.8-liter second-stage reactor, complete with Robinson-Mahoney internals, was assembled, pressure tested and installed downstream of the first-stage reactor. Some machine work was required to trim down oversized baffles on the catalyst basket in order to facilitate assembly of the 3.8-liter STR.

The new hydrogen compressor has not been installed as yet. The two-stage liquefaction experiments described in Task 7 were given priority over this activity due to time constraints associated with the contract schedule. It is anticipated that the compressor will be installed during the next phase of work.

TASK 5 - PREPARATION AND ANALYSIS OF MATERIALS

5.1 Coal Feedstocks

Two coal feedstocks were utilized during this phase of the project. The experimental program was principally focused on Lingan Harbour seam coal. Lingan and Prince mine coals were provided courtesy of the Cape Breton Development Corporation. Preparation involved riffling, grinding and screening. After screening, the individual coals were thoroughly mixed and stored in plastic-lined drums with a nitrogen blanket.

Proximate and ultimate analysis of the coals are given in Table 1. Proximate analysis, free-swelling index and forms of sulphur were determined according to standard methods. Total sulphur was measured using Eschka's method. Carbon, hydrogen and nitrogen were determined using a Perkin-Elmer elemental analyzer.

Petrographic analyses of the two coals are given in Table 2. These analyses were carried out by coal petrology experts at the Bedford Institute of Oceanography.

TABLE 1

ANALYSIS OF COALS TESTED

COAL DESCRIPTION:	LINGAN HR-6	PRINCE HB-D
Moisture, Wt. %	1.13	2.93
PROXIMATE (Dry Basis) Wt. %		
Ash	4.41	9.10
Volatile Matter	35.86	36.22
ULTIMATE (Dry Basis) Wt. %		
Carbon	81.07	73.48
Hydrogen	5.33	4.80
Nitrogen	1.46	1.45
Sulphur	0.96	4.22
Ash	4.41	9.10
Oxygen (by difference)	6.77	6.95
SULPHUR FORMS (Dry Basis) Wt. %		
Sulphate	0.01	0.18
Pyritic	0.47	2.25
Organic (by difference)	0.48	1.79
Total	0.96	4.22
FREE SWELLING INDEX	8½	2

TABLE 2

PETROGRAPHIC CHARACTERISTICS OF COALS TESTED

COAL DESCRIPTION:	LINGAN HR-6	PRINCE HB-D
MACERAL COMPOSITION (% by Volume)		
Vitrinite	78.8	74.7
Exinite	5.8	4.1
Inertinite	14.4	18.0
Macrinite	(5.8)	(0.8)
Micrinite	(2.2)	(5.4)
Semifusinite	(2.6)	(5.4)
Fusinite	(3.4)	(6.4)
Mineral Matter	1.0	3.2
Pyrite	(0.2)	(2.6)
Quartz	--	--
Shale	(0.8)	(0.4)
Calcite	--	(0.2)
% R_o Max.	1.01	0.78
V-Type Distribution, %		
V5	--	--
V6	--	6
V7	--	60
V8	4	26
V9	38	2
V10	54	6
V11	4	--

5.2 Anthracene Oil

This material is a coal-tar distillate which contains ca. 5.6 wt.% hydrogen. It was supplied by Domtar Chemicals Limited (Hamilton, Ontario). Hydrogenated solvents used in the two-stage liquefaction experiments were made from three batches of this highly aromatic oil.

The characteristics of raw oil are presented in Table 3. Carbon and hydrogen were determined using a Perkin-Elmer elemental analyzer. A standard kjeldahl method was used for the determination of nitrogen. Total sulphur content was measured by x-ray fluorescence. Oxygen was determined by neutron activation.

In this study, the hydrogen aromatic/aliphatic distribution was established by proton nuclear magnetic resonance (H-NMR). A Varian EM360L continuous wave spectrometer operating in the Fourier transform mode at 60 MHz, and a Nicolet 360 NB NMR spectrometer operating in the Fourier transform mode at 360 MHz were used to obtain H-NMR spectra. Proton assignments were as follows: aromatic hydrogens (H_{ar}) are found in the region of 9-6 ppm. Benzylic hydrogens, including methyls, occur at 3.5 to 2 ppm (H_{α}). Hydrogens, except methyls, on carbons one or more positions removed from aromatic rings, occur at 2-1.1 ppm (H_{β}). Aliphatic methyls are found at 1.1-0.4 ppm (H_{γ}). As almost no long aliphatic side chains are believed to be present, the H_{β} protons are predominantly in rings. These assignments are consistent with similar analyses reported in the literature (7,8). Figure 2 is a typical H-NMR spectrum of raw anthracene oil.

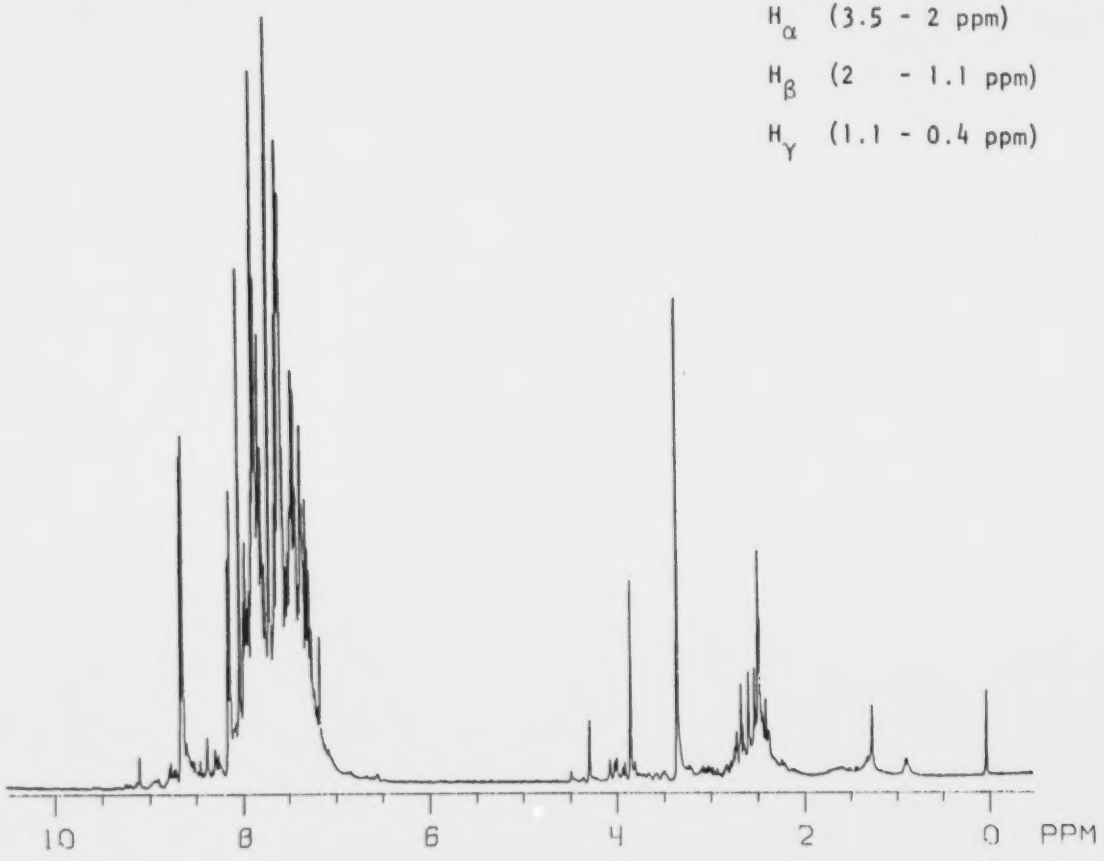
TABLE 3

ANALYSIS OF RAW ANTHRACENE OIL

SAMPLE DESCRIPTION:	BATCH NO.5-11	BATCH NO.5-12	BATCH NO.6-13
Specific Gravity, 60/60°F.	1.152	1.144	1.133
Carbon, Wt. %	91.67	90.93	91.10
Hydrogen, Wt. %	5.62	5.55	5.83
Nitrogen, Wt. %	0.88	1.01	0.91
Sulphur, Wt. %	0.56	0.56	0.62
Oxygen, Wt. %	1.21	0.54	0.71
Proton Distribution, %			
H_{ar} (9 - 6 ppm)	86.87	88.61	84.10
H_{α} (3.5 - 2 ppm)	10.74	8.97	11.00
H_{β} (2 - 1.1 ppm)	0.80	1.32	3.00
H_{γ} (1.1 - 0.4 ppm)	0.80	1.11	2.10
Distillation Data, °C. (ASTM D-1160)			
IBP	223	239	229
10 %	291	313	264
20 %	316	328	299
30 %	331	339	321
40 %	346	346	341
50 %	355	354	356
60 %	367	365	370
70 %	382	380	386
80 %	399	400	405
90 %	434	430	442
95 %	461	437	466
EP	482	490	489

Figure 2 - H-NMR SPECTRA OF RAW ANTHRACENE OIL WITH 5.62 WT. % HYDROGEN

PROTON DISTRIBUTION,	%
H_{ar} (9 - 6 ppm)	86.87
H_{α} (3.5 - 2 ppm)	10.74
H_{β} (2 - 1.1 ppm)	0:80
H_{γ} (1.1 - 0.4 ppm)	0.80



5.3 Catalysts

Two commercially available hydroprocessing catalysts, Katalco 504-K and Katalco 550 were evaluated in the two-stage coal liquefaction test program. The former catalyst is alumina-supported $NiO-MoO_3$ whereas the latter is alumina-supported $NiO-WO_3$. Both catalysts were used in extrudate form, 1.6 mm diameter. Nalcomo 479, an alumina-supported $CoO-MoO_3$ catalyst was used as received in the fixed-bed catalytic hydrotreater. The chemical and physical properties of these Ni/Mo, Ni/W and Co/Mo catalysts are given in Table 4. This data was provided by the manufacturers.

TABLE 4

CHEMICAL AND PHYSICAL PROPERTIES OF HYDROTREATING CATALYSTS

	KATALCO 504-K	KATALCO 550	NALCOMO 479
MoO_3, Wt. %	19.0		19.0
NiO, Wt. %	5.4	5.0	
CoO, Wt. %			4.4
WO_3, Wt. %		20.0	
Size, cm	0.16	0.16	0.16
Surface Area, M^2/g	170	230	180
Pore Volume, cc/g	0.43	0.50	0.47
Density, g/cc	0.86	0.79-0.81	0.82-0.84
Crush Strength, kg	8.6	6.4-7.3	6.4-7.3
Modal Pore Diameter, A°	100	100	100

Katalco 504-K and Katalco 550 are manufactured by the Katalco Corporation.

Nalcomo 479 is manufactured by the Nalco Chemical Company.

TASK 6 - HYDROGENATION OF ANTHRACENE OIL

Hydrogenated solvent used in Task 7 was prepared from raw anthracene oil using a fixed-bed catalytic hydrotreater. The bench-scale unit has been described in a previous report (3).

A commercial cobalt-molybdenum hydrotreating catalyst, Nalcomo 479, was used to prepare solvent containing 9% hydrogen. A summary of the operating conditions used to produce the hydrogenated oil is given in Table 5. The characteristics of two 9% H oil blends are shown in Table 6. An H-NMR spectrum of the 9% H solvent is given in Figure 3. Hydrogenated anthracene oil, 7.5% H, spiked with dimethyl disulphide was used to sulphide catalysts in Task 7. The 7.5% H oil was on hand from Phase V.

The two-stage coal liquefaction experiments conducted during Phase VI required in excess of 300 liters of hydrogenated solvent. The 9% H solvent was prepared at the rate of 1.8 liters per 7-hour man-day. Daily production was collected to obtain 20-liter batches. A total of sixteen 20-liter batches were prepared. Analysis of selected batches are presented in Table 7. Approximately eighty liters of this oil was used in commissioning the two-stage unit, exploratory trials and catalyst conditioning operations. Prior to and during the factorial test sequence, 120-liter blends were prepared by mixing six 20-liter batches. The analyses given in Table 6 are representative of the two 120-liter oil blends.

TABLE 5

REACTION CONDITIONS FOR ANTHRACENE OIL HYDROGENATION

Reactor Temperature °C.	363-380
Reactor Pressure, MPa	15.51
LHSV, hr^{-1}	0.20
Hydrogen Flow, L/hr	170-270
Product Hydrogen Content, Wt.%	9.0

Catalyst used: Nalcomo 479

TABLE 6

CHARACTERISTICS OF HYDROGENATED ANTHRACENE OIL

SAMPLE DESCRIPTION:	HAO 61	HAO 62
Specific Gravity, 60/60°F.	1.030	1.029
Carbon, Wt. %	90.26	90.27
Hydrogen, Wt. %	9.03	9.01
Nitrogen, Wt. %	0.10	0.12
Sulphur, Wt. %	0.007	0.008
Oxygen, Wt. %	0.20	0.20
Proton Distribution, %		
H_{ar} (9 - 6 ppm)	27.78	26.46
H_{α} (3.5 - 2 ppm)	26.25	27.75
H_{β} (2 - 1.1 ppm)	35.68	36.15
H_{γ} (1.1 - 0.4 ppm)	10.29	9.65
Distillation Data, °C (ASTM D-1160)		
IBP	191	186
10 %	257	255
20 %	280	277
30 %	297	293
40 %	308	304
50 %	318	314
60 %	328	322
70 %	342	335
80 %	363	353
90 %	390	379
95 %	416	399
EP	437	433

Figure 3 - H-NMR SPECTRA OF HYDROGENATED ANTHRACENE OIL WITH 9.01 WT. % HYDROGEN

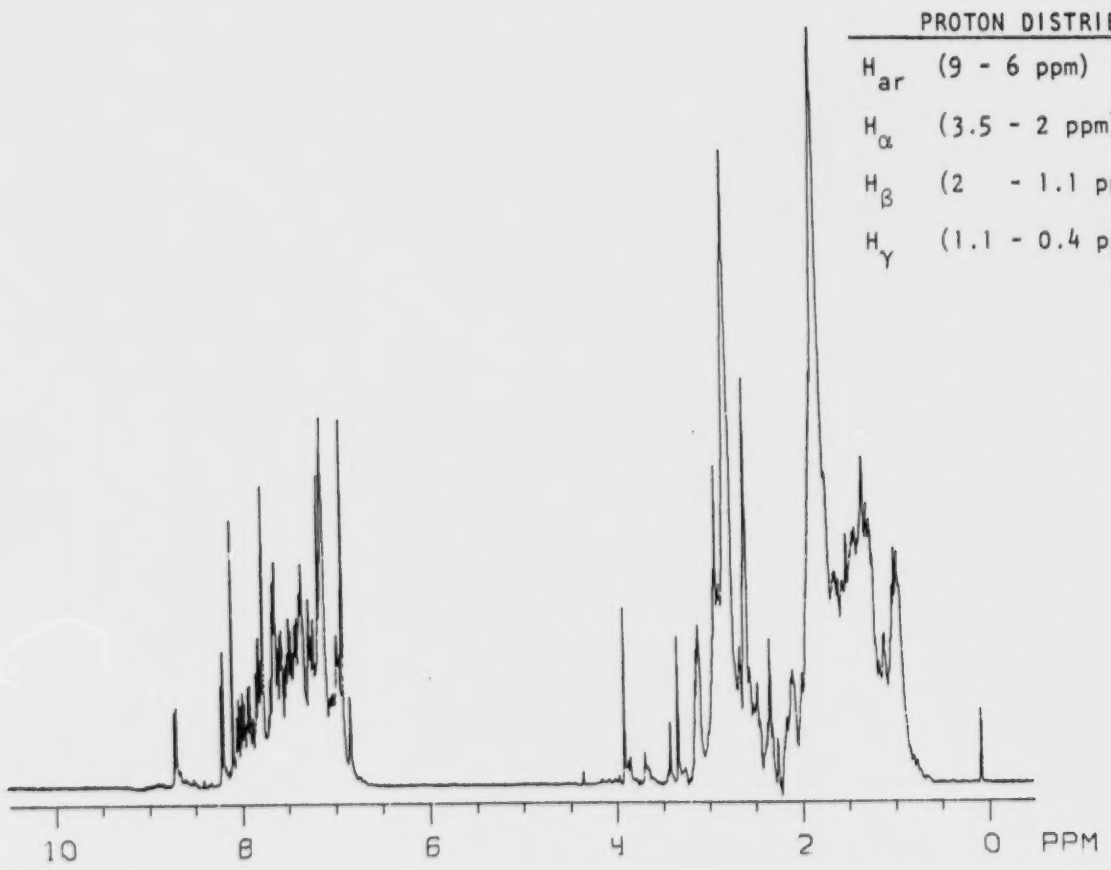


TABLE 7

ANALYSIS OF 20-LITER BATCHES OF 9% H SOLVENT

BATCH NO.	1	2	3	4	5	6	7	8
Specific Gravity, 60/60°F	1.027	1.027	1.027	1.029	1.029	1.032	1.031	1.030
Carbon, Wt. %	90.27	89.96	89.92	89.40	89.49	89.24	89.64	89.80
Hydrogen, Wt. %	9.23	9.20	9.22	9.11	9.10	8.99	9.10	8.96
Nitrogen, Wt. %	0.10	0.13	0.14	0.13	0.15	0.13	0.12	0.13
Sulphur, Wt. %	0.006	0.008	0.009	0.010	0.009	0.007	0.008	0.008
Proton Distribution, %								
H_{ar} (9 - 6 ppm)	27.39		25.36		25.51			26.46
H_{α} (3.5 - 2 ppm)	24.42		25.07		27.00			27.07
H_{β} (2 - 1.1 ppm)	38.28		39.65		37.98			34.85
H_{γ} (1.1 - 0.4 ppm)	9.90		9.91		9.50			7.06
Distillation Data, °C (ASTM D-86)								
IBP	175	163	167	172	179	175	185	180
5 %	242	241	243	249	242	239	250	250
10 %	263	265	268	259	262	266	267	267
20 %	283	284	285	287	280	284	286	290
30 %	297	295	298	298	297	297	300	301
40 %	307	306	307	308	309	308	310	311
50 %	317	316	317	318	319	318	320	321
60 %	327	326	326	327	328	328	330	330
70 %	340	338	338	338	340	340	340	342
80 %	358	355	355	354	354	357	357	358
90 %	387	383	381	382	380	380	381	382
95 %	418	413	404	406	400	397	404	409
EP	495	467	468	488	460	463	495	530

TABLE 7 (CONTINUED)

ANALYSIS OF 20- LITER BATCHES OF 9% H SOLVENT

BATCH NO.	9	10	11	12	13	14	15	16
Specific Gravity, 60/60°F.	1.030	1.031	1.031	1.030	1.027	1.031	1.028	1.026
Carbon, Wt. %	88.73	88.92	89.85	88.74	89.18	89.45	89.93	90.63
Hydrogen, Wt. %	9.07	9.06	8.97	8.97	9.06	8.97	8.90	9.10
Nitrogen, Wt. %	0.14	0.12	0.10	0.10	0.11	0.16	0.11	0.09
Sulphur, Wt. %	0.008	0.005	0.007	0.009	0.012	0.016	0.013	0.010
Proton Distribution, %								
H_{ar} (9 - 6 ppm)	26.00			25.79		28.72		25.80
H_{α} (3.5 - 2 ppm)	27.23			27.27		27.96		26.17
H_{β} (2 - 1.1 ppm)	37.92			37.80		34.99		37.64
H_{γ} (1.1 - 0.4 ppm)	8.85			9.14		8.32		10.38
Distillation Data, °C (ASTM D-86)								
IBP	188	184	184	194	162	170	170	169
5 %	249	245	246	257	240	236	235	245
10 %	271	266	263	269	258	255	252	261
20 %	290	286	282	288	277	276	270	279
30 %	301	300	294	299	292	292	288	293
40 %	311	310	307	308	306	304	302	305
50 %	321	320	317	318	315	315	312	315
60 %	331	330	327	326	325	325	322	326
70 %	343	342	338	338	337	337	334	336
80 %	360	357	354	353	352	353	350	352
90 %	386	384	380	377	377	376	371	375
95 %	414	413	405	395	400	397	389	398
EP	535	520	482	495	465	493	465	476

TASK 7 - TWO-STAGE THERMAL/CATALYTIC LIQUEFACTION

The purpose of this task was to initiate experimental studies on integrated two-stage thermal/catalytic liquefaction applied to Sydney area coals. In this process configuration, coal is solubilized in the thermal first stage and subsequently upgraded in the catalytic second stage. By using two separate reaction zones, the thermal dissolution and the catalytic upgrading steps can be independently optimized to maximize efficiency and control product distribution and quality. Thus, the two-stage processing scheme offers the potential of higher distillate yields, improved hydrogen utilization and reduced production of hydrocarbon gases.

7.1 Commissioning Trials and Exploratory Tests

Assembly of the integrated two-stage liquefaction unit was completed around May 15, 1985. The second-stage reactor was charged with 450 g of Katalco 504-K nickel-molybdenum catalyst. Following a series of pressure tests with argon and hydrogen, one run was carried out with oil alone at room temperature and 17.24 MPa. Prior to initiating coal liquefaction experiments, the catalyst was sulphided using hydrogenated anthracene oil, 7.5% H, spiked with dimethyl disulphide. Catalyst sulphiding operations were conducted at 17.24 MPa and a start-of-run temperature of 180°C. The catalyst was sulphided over a 16-hour period. Reactor temperature was increased at the rate of about 12°C/hour to a maximum of 370°C. "Coal-in" operations began in June with Lingan Harbour seam coal.

Three commissioning trials (Run Nos. 6-CT-1, 6-CT-2 and 6-CT-3) were conducted at 17.24 MPa varying first- and second-stage reactor temperatures, slurry space velocity and hydrogen treatrate. Test results are shown in Table 8. Detailed performance and material balance data are presented in Appendices B and C. High coal conversions were achieved in all three runs, ca. 93%. Oil yields ranged from 44 to 49% based on moisture ash-free coal. It should be noted at this point, however, that in Run No. 6-CT-1 the unit was on-stream with oil alone for 3.5 hours prior to feeding coal/oil slurry. Furthermore, approximately 1.5 kg of hydrogenated anthracene oil was left behind in the catalytic reactor after the flush cycle in Run Nos. 6-CT-1 and 6-CT-2. The presence of oil in the system at start-up would bias coal conversion and oil yield high in runs of short duration.

After completion of the commissioning trials, it was decided to conduct four additional runs to further explore operating conditions. The runs were carried out varying unit back pressure, first-stage reactor temperature and slurry space velocity. Results of experimental tests are presented in Table 8. The highest oil yield, 50.7 wt.% maf coal, was achieved in Run No. 6002 conducted at 20.68 MPa with thermal and catalytic reactor temperatures of 450 and 425°C, respectively. In Run No. 6002, the C_4^+ oil yield is estimated to be about 60% based on maf coal plus hydrogen added.

Test results from the commissioning trials and exploratory runs were used to select the operating conditions for the factorial test sequence presented hereafter.

TABLE 8

RESULTS OF PRELIMINARY TWO-STAGE COAL LIQUEFACTION TRIALS

RUN NO.	6-CT-1	6-CT-2	6-CT-3	6001	6002	6003	6004
Temperatures: °C							
Thermal Dissolver	460	460	450	450	450	460	440
Catalytic Reactor	400	400	425	425	425	425	425
Back Pressure, MPa	17.24	17.24	17.24	17.24	20.68	20.68	20.68
Slurry Feedrate, kg/hr	1.329	0.958	0.971	1.961	0.985	0.988	0.971
H_2 Treatrate, Wt. % Dry Coal	11.4	12.3	15.1	12.0	15.0	15.0	15.2
Catalyst Age (kg coal/kg catalyst)	6	11	17	28	34	39	45
CONVERSIONS, WT. % OF maf COAL							
Pentane Solubles plus Gases	66.6	70.1	74.1	53.8	73.8	73.8	68.3
Toluene Solubles plus Gases	85.5	88.0	89.7	76.2	87.6	88.0	84.9
THF Solubles plus Gases	92.8	93.4	93.5	88.7	92.2	92.0	91.3
PRODUCT DISTRIBUTION, WT. % OF maf COAL							
Gases	20.2	26.5	24.8	16.4	23.1	29.2	20.5
Oil	46.4	43.6	49.3	37.4	50.7	44.6	47.8
Asphaltenes	18.9	17.9	15.6	22.4	13.8	14.2	16.6
Preasphaltenes	7.3	5.4	3.8	12.5	4.6	4.0	6.4
H_2 CONSUMPTION, WT. % OF maf COAL	5.9	6.2	6.3	5.0	7.3	7.3	7.2

7.2 Process Variable Study on Lingan Harbour Seam Coal

A series of sixteen experiments were carried out according to a factorial design to evaluate the effects of second-stage reactor temperature, operating pressure, slurry space velocity and catalyst on raw product yields from Lingan Harbour seam coal. Commercial hydroprocessing catalysts, Katalco 504-K and Katalco 550, were used in this study. Katalco 504-K is an alumina-supported nickel-molybdenum catalyst whereas Katalco 550 is alumina-supported nickel-tungsten. Both catalysts have unimodal pore size distributions with pores of about 100 angstroms diameter.

Hydrogenated anthracene oil containing 9% H was used as vehicle. The concentration of coal in the feed slurry was fixed at 27 wt.%. Hydrogen treatrate was ca. 15 wt.% based on dry coal. The operating conditions investigated were as follows:

	Thermal Stage	Catalytic Stage
Reactor Temperature, °C	450	400, 425
SHSV, Hr^{-1}	1.00, 1.50	0.33, 0.50
Back Pressure, MPa	17.24, 20.68	17.24, 20.68

A typical two-stage liquefaction run was of 8-10 hours duration. However, an additional 5-6 hours was required for preliminary and post-run operations. In the majority of runs, the controlled variables lined-out about 4 hours after start-up. During the steady state operating period, vent gas streams were sampled and other product streams were collected for analytical and material balance purposes. A detailed operating procedure is given in Appendix F.

In these experiments, the following definitions for the product fractions isolated by solvent extraction were used:

oil - pentane solubles

asphaltenes - toluene solubles, pentane insolubles

preasphaltenes - THF solubles, toluene insolubles.

A direct extraction technique was used to determine these fractions. Individual samples of product slurry were separately extracted with pentane, toluene and tetrahydrofuran (THF). Conversions were calculated on the basis of maf coal fed to the reaction zone during the steady-state sampling period and unconverted coal in the dry insoluble residues. Conversion calculations are described in Appendix A.

Vent gas streams were analyzed using a Perkin-Elmer Sigma 3B gas chromatograph interfaced with a Perkin-Elmer Sigma 10 Chromatography data station. Simulated distillations were also done with this instrumentation using the D-2887 Sigma Basic Program.

Results from the sixteen two-stage thermal/catalytic liquefaction runs are summarized in Table 9. An examination of the data indicates that the controlled variables do affect product distribution. A statistical evaluation of the data was carried out to determine the significance of the observed effects. The response variables selected for the statistical analysis included gas make, oil yield, asphaltenes, preasphaltenes and hydrogen consumption. A standard analysis of variance was used to establish confidence levels. Details are presented in Appendix E.

TABLE 9

EVALUATION OF PROCESS OPERATING CONDITIONS WITH LINGAN COAL

RUN NO.	6010	6008	6012	6006	6011	6005	6007	6009
Temperatures: °C								
Thermal Dissolver	450	450	450	450	450	450	450	450
Catalytic Reactor	400	400	400	400	425	425	425	425
Back Pressure, MPa	20.68	17.24	20.68	17.24	20.68	17.24	20.68	17.24
Slurry Feedrate, kg/hr	1.002	0.986	1.480	1.462	1.007	0.993	1.466	1.470
H_2 Treatrate, Wt. % Dry Coal	14.7	15.0	15.0	15.2	14.9	14.9	15.1	15.1
Catalyst Age (kg coal/kg catalyst)	90	74	106	59	97	51	69	83
CONVERSIONS, WT. % OF maf COAL								
Pentane Solubles plus Gases	61.2	58.6	50.5	49.0	68.9	67.1	62.4	59.8
Toluene Solubles plus Gases	81.9	80.7	74.5	73.6	85.3	84.9	82.3	81.2
THF Solubles plus Gases	90.5	90.0	88.6	88.4	91.2	91.0	90.4	90.0
PRODUCT DISTRIBUTION, WT. % OF maf COAL								
Gases	20.6	20.6	15.7	16.8	23.4	24.5	19.6	21.7
Oil	40.6	38.0	34.8	32.2	45.5	42.6	42.8	38.1
Asphaltenes	20.7	22.1	24.0	24.6	16.4	17.8	19.9	21.4
Preasphaltenes	8.6	9.3	14.1	14.8	5.9	6.1	8.1	8.8
H_2 CONSUMPTION, WT. % OF maf COAL	5.6	5.2	5.1	4.7	6.7	5.6	6.3	5.3

Catalyst tested: Katalco 504-K
Detailed product distribution given in Appendix B

TABLE 9 (CONTINUED)

EVALUATION OF PROCESS OPERATING CONDITIONS WITH LINGAN COAL

RUN NO.	6025	6022	6023	6020	6019	6018	6024	6021
Temperatures: °C								
Thermal Dissolver	450	450	450	450	450	450	450	450
Catalytic Reactor	400	400	400	400	425	425	425	425
Back Pressure, MPa	20.68	17.24	20.68	17.24	20.68	17.24	20.68	17.24
Slurry Feedrate, kg/hr	1.001	1.037	1.489	1.517	1.078	0.989	1.523	1.489
H_2 Treatrate, Wt. % Dry Coal	14.7	14.2	14.9	14.7	14.1	14.9	14.6	14.9
Catalyst Age (kg coal/kg catalyst)	97	73	81	54	45	38	91	67
CONVERSIONS, WT. % OF maf COAL								
Pentane Solubles plus Gases	57.3	53.3	48.7	46.6	69.4	67.7	57.1	56.7
Toluene Solubles plus Gases	79.8	78.1	73.6	71.3	85.3	85.0	79.4	78.7
THF Solubles plus Gases	90.3	89.6	88.8	87.9	91.8	91.3	89.4	89.0
PRODUCT DISTRIBUTION, WT % OF maf COAL								
Gases	16.5	17.8	14.4	15.2	23.3	23.6	18.4	19.1
Oil	40.8	35.5	34.3	31.4	46.1	44.1	38.7	37.6
Asphaltenes	22.5	24.8	24.9	24.7	15.9	17.3	22.3	22.0
Preasphaltenes	10.5	11.5	15.2	16.6	6.5	6.3	10.0	10.3
H_2 CONSUMPTION, WT % OF maf COAL	5.3	4.2	5.2	4.2	6.0	6.0	5.7	5.4

Catalyst tested: Katalco 550
Detailed product distribution given in Appendix B

Temperature in the second stage was found to have a significant effect on product distribution and hydrogen consumption. Increasing the temperature of the catalytic reactor from 400 to 425°C affected an increase in hydrogen consumption, higher yields of gases and oil and lower yields of asphaltenes and preasphaltenes. A comparison of Run No. 6010 at 400°C and Run No. 6011 at 425°C shows the following shift in response variables:

- gases increased from 20.6 to 23.4%
- oil increased from 40.6 to 45.5%
- asphaltenes decreased from 20.7 to 16.4%
- preasphaltenes decreased from 8.6 to 5.9%
- hydrogen consumption increased from 5.6 to 6.7%.

The effect of pressure was significant with regard to gas make, oil yield, asphaltene yield and hydrogen consumption. Increasing the operating pressure from 17.24 to 20.68 MPa had a positive effect on oil yield and hydrogen consumption and a negative effect on gas make and asphaltene yield. A comparison of Run No. 6009 at 17.24 MPa with Run No. 6007 at 20.68 MPa illustrates the directional trends in these response variables. Results were as follows:

- gases decreased from 21.7 to 19.6%
- oil increased from 38.1 to 42.8%
- asphaltenes decreased from 21.4 to 19.9%
- hydrogen consumption increased from 5.3 to 6.3%.

Slurry feedrate, and consequently slurry space velocity, was found to significantly affect all five response variables. Increasing the

slurry feedrate from 1 to 1.5 kg/hr affected a decrease in hydrogen consumption, lower yields of gases and oil and higher yields of asphaltenes and preasphaltenes. A comparison of Run No. 6010 at 1 kg/hr with Run No. 6012 at 1.5 kg/hr shows the following shift in response variables:

- gases decreased from 20.6 to 15.7%
- oil decreased from 40.6 to 34.8%
- asphaltenes increased from 20.7 to 24.0%
- preasphaltenes increased from 8.6 to 14.1%
- hydrogen consumption decreased from 5.6 to 5.1%.

In general, coal conversions and product distributions were quite similar for both catalysts tested under identical operating conditions. The statistical evaluation, however, shows that the lower hydrogen consumptions and gas makes observed with Katalco 550 are significant. Furthermore, the lower yields of preasphaltenes with Katalco 504-K are also significant. A comparison of data from Run No. 6010 with Katalco 504-K and Run No. 6025 with Katalco 550 illustrate the directional trends observed. In Run No. 6010 hydrogen consumption, gas make and preasphaltenes were 5.6, 20.6 and 8.6%, respectively, as compared with 5.3, 16.5 and 10.5% in Run No. 6025. Oil yield in both cases was ca. 41%.

Duplicate runs were conducted with aged Katalco 504-K catalyst to assess the reliability of test data. Run No. 6014 (catalyst age: 114 kg coal/kg catalyst) was carried out at operating conditions similar to Run No. 6011 (catalyst age: 97 kg coal/kg catalyst). Raw-product distribution and hydrogen consumption were essentially the same for both experiments as shown in Table 10. It should be noted at this point that the catalyst

TABLE 10

RAW PRODUCT DISTRIBUTION AS A FUNCTION OF CATALYST TYPE AND AGE

RUN NO. COAL TESTED CATALYST TESTED	6002 Lingan HR-6 Katalco 504-K	6011 Lingan HR-6 Katalco 504-K	6014 Lingan HR-6 Katalco 504-K	6019 Lingan HR-6 Katalco 550	6026 Lingan HR-6 Katalco 550
Temperatures: °C					
Thermal Dissolver	450	450	450	450	450
Catalytic Reactor	425	425	425	425	425
Back Pressure, MPa	20.68	20.68	20.68	20.68	20.68
Slurry Feedrate, kg/hr	0.985	1.007	0.984	1.078	1.055
H_2 Treatrate, Wt. % Dry Coal	15.0	14.9	15.0	14.1	14.0
Catalyst Age (kg coal/kg catalyst)	34	97	114	45	105
CONVERSIONS, WT. % OF maf COAL					
Pentane Solubles plus Gases	73.8	68.9	69.5	69.4	68.8
Toluene Solubles plus Gases	87.6	85.3	85.9	85.3	84.8
THF Solubles plus Gases	92.2	91.2	91.0	91.8	91.9
PRODUCT DISTRIBUTION, WT. % OF maf COAL					
Gases	23.1	23.4	24.1	23.3	21.8
Oil	50.7	45.5	45.4	46.1	47.0
Asphaltenes	13.8	16.4	16.4	15.9	16.0
Preasphaltenes	4.6	5.9	5.1	6.5	7.1
H_2 CONSUMPTION, WT. % OF maf COAL	7.3	6.7	6.9	6.0	5.6

exhibited a higher level of activity at an earlier age. The observed decline in initial activity, however, is not necessarily indicative of the overall rate of catalyst deactivation.

In the studies conducted with Katalco 550, catalyst activity was also investigated as a function of age. Run No. 6026 (catalyst age: 105 kg coal/kg catalyst) was carried out at the same operating conditions as Run No. 6019 (catalyst age: 45 kg coal/kg catalyst). Raw-product distributions and hydrogen consumption were very similar in both cases as shown in Table 10. These results suggest that catalyst activity was fairly constant throughout the duration of the test period.

Analysis of pentane, toluene and THF-soluble fractions from these experiments are presented in Tables 11 and 12. Analysis of distillate product from flashed separator bottoms of selected runs are given in Table 13. Examination of the elemental analyses and the H-NMR data indicates that the Katalco 504-K catalyst is more active than Katalco 550 for hydrogenation, denitrogenation and deoxygenation reactions. Work on filtered separator bottoms product suggests that Katalco 550 has superior cracking ability - further investigation is warranted.

TABLE 11

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM TWO-STAGE LIQUEFACTION TESTS

RUN NO.	6005	6006	6007	6008	6009	6010	6011	6012	6014
Pentane Soluble Fraction									
Carbon, Wt. %	87.99	90.59	89.38	88.79	89.93	90.35	90.14	89.55	89.85
Hydrogen, Wt. %	9.12	9.02	9.33	9.04	9.05	9.34	9.13	9.16	9.11
Nitrogen, Wt. %	0.14	0.14	0.15	0.17	0.15	0.16	0.14	0.15	0.15
Sulphur, Wt. %	0.007	0.010	0.012	0.009	0.007	0.011	0.011	0.011	0.015
Toluene Soluble Fraction									
Carbon, Wt. %	88.36	90.30	90.26	90.01	90.31	90.31	90.45	89.95	89.79
Hydrogen, Wt. %	8.92	8.88	9.07	8.83	8.83	9.11	9.06	8.78	8.85
Nitrogen, Wt. %	0.22	0.28	0.22	0.28	0.26	0.25	0.22	0.26	0.20
Sulphur, Wt. %	0.012	0.019	0.017	0.015	0.016	0.018	0.015	0.017	0.019
THF Soluble Fraction									
Carbon, Wt. %	88.96	90.26	90.63	90.71	89.45	88.73	89.45	88.69	88.97
Hydrogen, Wt. %	8.84	8.94	9.06	8.82	8.94	8.98	9.04	8.67	9.05
Nitrogen, Wt. %	0.23	0.29	0.22	0.28	0.25	0.26	0.21	0.26	0.24
Sulphur, Wt. %	0.011	0.019	0.015	0.016	0.017	0.016	0.012	0.019	0.016

TABLE 11 (CONTINUED)

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM TWO-STAGE LIQUEFACTION TESTS

RUN NO.	6018	6019	6020	6021	6022	6023	6024	6025	6026	6027
Pentane Soluble Fraction										
Carbon, Wt. %	90.36	89.53	90.88	89.38	88.75	89.84	89.50	88.22	88.50	88.45
Hydrogen, Wt. %	9.04	9.26	9.02	8.97	8.85	8.59	8.78	8.73	9.04	8.89
Nitrogen, Wt. %	0.21	0.19	0.19	0.20	0.21	0.20	0.20	0.18	0.19	0.17
Sulphur,Wt. %	0.009	0.011	0.011	0.009	0.009	0.013	0.012	0.016	0.011	0.031
Toluene Soluble Fraction										
Carbon, Wt. %	89.84	89.79	90.19	89.51	88.64	89.66	89.52	88.25	88.46	87.92
Hydrogen, Wt. %	8.69	8.86	8.66	8.53	8.75	8.41	8.75	8.68	8.74	8.62
Nitrogen, Wt. %	0.28	0.27	0.30	0.30	0.33	0.32	0.28	0.31	0.28	0.22
Sulphur, Wt. %	0.014	0.017	0.022	0.020	0.018	0.025	0.021	0.026	0.023	0.041
THF Soluble Fraction										
Carbon, Wt. %	88.55	89.58	89.14	88.90	88.87	88.23	88.20	87.30	88.06	87.45
Hydrogen, Wt. %	8.74	8.89	8.61	8.62	8.57	8.30	8.70	8.66	8.82	8.72
Nitrogen, Wt. %	0.28	0.26	0.32	0.32	0.33	0.32	0.30	0.29	0.29	0.23
Sulphur, Wt. %	0.016	0.015	0.025	0.019	0.022	0.023	0.019	0.027	0.11	0.037

TABLE 12

DISTILLATION DATA ON PENTANE SOLUBLE FRACTIONS
FROM TWO-STAGE COAL LIQUEFACTION TESTS

RUN NO.	6006	6007	6011	6019	6020	6027
Distillation Data, °C (by Gas Chromatography)						
Pentane Soluble Fraction						
IBP/5	160/209	149/208	143/208	143/207	168/209	154/207
10/30	237/282	232/279	232/279	227/274	236/283	229/279
50	311	308	308	306	311	307
70/90	339/383	336/381	336/381	333/378	340/384	336/381
95/EP	412/476	412/478	412/478	408/478	413/478	412/478

TABLE 13

ANALYSIS OF DISTILLATE PRODUCT
FROM SEPARATOR BOTTOMS

RUN NO.	6011	6019	6027
Specific Gravity, 60/60 °F	1.006	1.011	1.018
Viscosity, cst @ 40°C	6.05	5.64	6.52
Conradson Carbon, Wt. %	0.23	0.17	0.32
Carbon, Wt. %	89.08	88.13	88.02
Hydrogen, Wt. %	9.22	9.13	9.30
Nitrogen, Wt. %	0.10	0.18	0.12
Sulphur, Wt. %	0.006	0.006	0.005
Oxygen, Wt. %	0.17	0.23	0.25
Proton, Distribution, %			
H_{ar} (9 - 6 ppm)	23.81	26.18	27.40
H_{α} (3.5 - 2 ppm)	25.40	23.53	24.20
H_{β} (2 - 1.1 ppm)	38.10	35.18	33.81
H_{γ} (1.1 - 0.4 ppm)	12.70	15.12	14.59
Distillation Data, °C (ASTM D-1160)			
IBP	144	151	170
10 %	241	236	241
20 %	263	261	266
30 %	279	278	284
40 %	291	291	298
50 %	304	304	310
60 %	317	317	322
70 %	327	332	340
80 %	358	357	366
90 %	397	395	403
95 %	440	437	457
EP	462	470	496

7.3 Comparison of Lingan Harbour and Prince Hub Seam Coal Reactivity

After completion of experimental studies on Lingan Harbour seam coal with the unimodal micropore catalysts, one run was conducted with Prince Hub seam coal. The run was made with Katalco 550 since the second-stage reactor was charged with that catalyst. Temperature in the thermal first-stage reactor was reduced to 440°C. This temperature was selected based on our earlier operating experience with Prince mine coal. Test results are presented in Table 14.

Examination of the data shows a high level of coal conversion, 92.9%. Run No. 6027 with Prince coal produced a higher C_4^+ liquid yield and lighter products than Run No. 6026 with Lingan Coal. The C_4^+ liquid yield in the former case was 65.7% compared with 61.9% in the latter. C_4^+ liquid yields were calculated on the basis of C_4^- gas make plus light overhead organic liquids and separator bottoms liquid recovered by filtration at 75°C.

The product distributions shown in Figure 4 illustrate that Prince Hub seam coal is more reactive than Lingan Harbour seam coal. These results are in agreement with trends observed earlier under donor solvent and catalytic liquefaction conditions. The higher reactivity of Hub seam coal is primarily attributed to its lower rank.

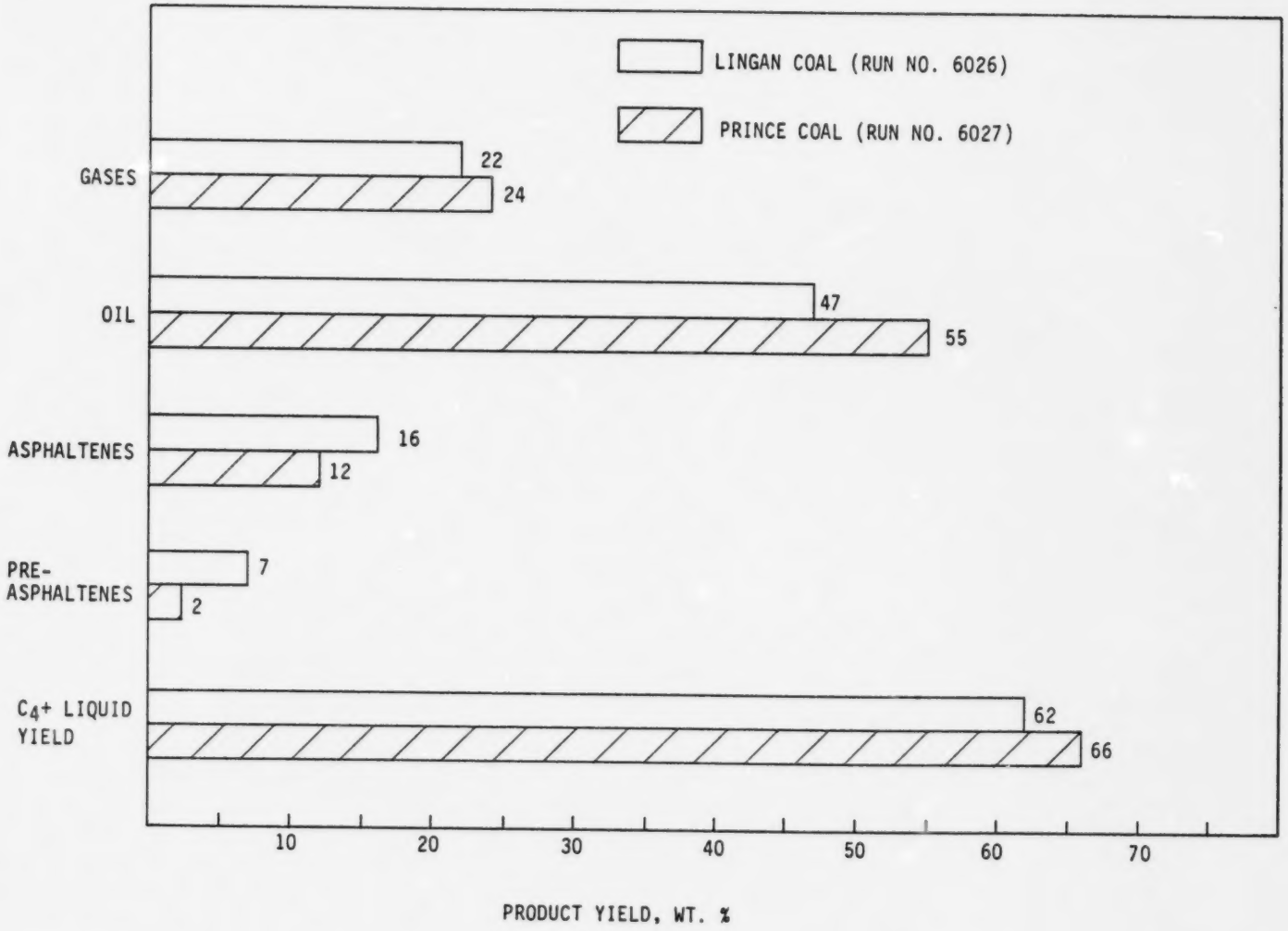
TABLE 14

CONVERSION AND PRODUCT DISTRIBUTION DATA FOR
LINGAN HARBOUR AND PRINCE HUB SEAM COALS

	LINGAN HR-6 RUN NO. 6026	PRINCE HB-D RUN NO. 6027
Temperatures: °C		
Thermal Dissolver	450	440
Catalytic Reactor	425	425
Back Pressure, MPa	20.68	20.68
Slurry Feedrate, kg/hr	1.055	1.101
H_2 Treatrate, Wt. % Dry Coal	14.0	14.6
Catalyst Age (kg coal/kg catalyst)	105	111
CONVERSIONS, Wt % OF maf COAL		
Pentane Solubles plus Gases	68.8	78.8
Toluene Solubles plus Gases	84.8	90.9
THF Solubles plus Gases	91.9	92.9
PRODUCT DISTRIBUTION, WT. % OF maf COAL		
Gases	21.8	23.5
Oil	47.0	55.3
Asphaltenes	16.0	12.1
Preasphaltenes	7.1	2.0
C_4 Liquid Yield, Wt. %*	61.9	65.7
H_2 CONSUMPTION, WT. % OF maf COAL	5.6	6.2

* Based on maf coal plus hydrogen added
Catalyst used: Katalco 550

Figure 4 - PRODUCT DISTRIBUTION AS A FUNCTION OF COAL FEEDSTOCK



CONCLUSIONS AND RECOMMENDATIONS

Phase VI of a research program on the direct hydroliquefaction of Nova Scotia coals has been completed. Our bench-scale coal liquefaction unit has been modified and expanded for two-stage operations. Process studies have been initiated using unimodal micropore catalysts in the second-stage reactor.

A total of twenty-five liquefaction experiments was conducted during this phase of work with Lingan Harbour seam coal. The runs were made in a once-through mode using hydrogenated anthracene oil, 9% H content, as reaction solvent. As a result of the test work, it was found that:

(a) Catalytic reactor temperature, over the range 400 to 425°C, has a significant effect on product composition and hydrogen consumption. In general, higher gas makes and oil yields were observed at 425°C.

(b) Pressure, over the range 17.24-20.68 MPa, had little effect on coal conversion. However, increasing the operating pressure had a positive effect on oil yield and hydrogen consumption.

(c) Slurry feedrate, over the range 1-1.5 kg/hr affected product yields and hydrogen consumption. Increasing the residence time of reacting coal fragments in the reaction zone resulted in higher yields of gases and

oil and lower yields of asphaltenes and preasphaltenes.

(d) Both catalysts tested, Katalco 504-K nickel-molybdenum and Katalco 550 nickel-tungsten, produced high coal conversions, ca. 90%. The former catalyst exhibited a higher degree of activity for hydrogenation, denitro genation and deoxygenation than the latter. Work on separator bottoms product suggests that Katalco 550 has superior cracking ability - further investigation is warranted.

Duplicate runs were carried out with both catalysts to assess the reliability of test data. Katalco 504-K showed a slightly higher activity at an earlier age. However, experimental results indicate that the activity of both catalysts remained fairly constant throughout the test period.

Two-stage thermal/catalytic operations with Prince Hub seam coal produced higher C_4^+ liquid yield and lighter products than operations with Lingan Harbour seam coal. Maximum C_4^+ liquid yields, to date, from Prince and Lingan coals were 66 and 62 wt.%, respectively, based on maf coal plus hydrogen added. In the former case, 55 wt.% of the maf coal was converted to pentane soluble oil compared with 47 wt.% in the latter. The higher reactivity of Hub seam coal is primarily attributed to its lower rank.

Based on the results of this work, the following recommendations for future work are proposed:

(a) Conduct additional test work on Prince Hub seam coal with Katalco 550 catalyst to further investigate product distribution as a function of thermal dissolver temperature and slurry space velocity.

(b) Determine the reactivity of Donkin-Morien Harbour seam coal with the unimodal micropore catalyst mentioned above.

(c) Evaluate the performance of at least one bimodal catalyst in the second-stage reactor.

(d) On completion of the thermal/catalytic liquefaction studies, convert the thermal dissolver to a catalytic dissolver. Following this, investigate the potential of catalytic two-stage processing applied to Sydney area coals.

(e) Finally, with the most promising combination of coal and process configuration, conduct longer term runs with liquid recycle to determine ultimate yields, product composition and catalyst aging characteristics.

At present, conventional coal liquefaction is not commercially viable in Eastern Canada. Process developments are required to improve distillate yield and/or reduce plant capital costs. Two-stage direct liquefaction

offers the potential to enhance distillate yield and improve hydrogen utilization. Extensive test work is required on two-stage process configurations to determine the extent to which these improvements can be realized.

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APPENDIX A

CONVERSION CALCULATIONS

APPENDIX A

CONVERSION CALCULATIONS FOR CONTINUOUS COAL LIQUEFACTION EXPERIMENTS

Calculations are based on operations during the steady state sampling period.

CONVERSIONS, WT.%:

$$\text{Pentane sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic pentane insol.})}{\text{maf coal charged}} \times 100$$

$$\text{Toluene sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic toluene insol.})}{\text{maf coal charged}} \times 100$$

$$\text{THF sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic THF insol.})}{\text{maf coal charged}} \times 100$$

$$\text{Gases} = \frac{\text{product gases}}{\text{maf coal charged}} \times 100$$

DEFINITION OF PRODUCTS

Oil = pentane solubles

Asphaltenes = toluene solubles - pentane solubles

Preasphaltenes = THF solubles - toluene solubles

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6-CT-1	6-CT-2	6-CT-3
COAL TESTED	Lingan L	Lingan L	Lingan L
CATALYST TESTED	Katalco 504-K	Katalco 504-K	Katalco 504-K
A. OPERATING CONDITIONS			
Unit back pressure, MPa	17.24	17.24	17.24
Temperatures, °C:			
Thermal Dissolver	460	460	450
Catalytic Reactor	400	400	425
Slurry Preheater	173	238	236
Hot Separator	300	300	300
Feedrates:			
Slurry, kg/hr	1.329	0.958	0.971
Raw Coal, kg/hr	0.332	0.240	0.243
Hydrogen, L/hr	414.	324.	402.
SHSV, hr^{-1}; Thermal Dissolver	1.35	1.00	1.00
Catalytic Reactor	0.45	0.33	0.33
Hydrogen Treatrate, Wt. % Dry Coal	11.4	12.3	15.1
Catalyst Age (kg coal/kg catalyst)	6	11	17
B. CONVERSIONS OF maf COAL, WT. %			
Pentane Solubles plus Gases	66.6	70.1	74.1
Toluene Solubles plus Gases	85.5	88.0	89.7
THF Solubles plus Gases	92.8	93.4	93.5
C. PRODUCT YIELDS ON maf COAL, WT. %			
CH_4	4.3	5.7	4.9
C_2H_6	3.8	4.9	4.9
C_3H_8	3.2	4.5	3.6
C_4H_{10}	2.9	4.0	4.1
CO	0.2	0.2	0.1
H_2S	0.3	0.1	0.1
H_2O	5.4	6.9	6.9
NH_3	0.1	0.2	0.2
Oil	46.4	43.6	49.3
Asphaltenes	18.9	17.9	15.6
Preasphaltenes	7.3	5.4	3.8
Unreacted Coal	7.2	6.6	6.5
D. H_2 CONSUMPTION, WT. % OF maf COAL	5.9	5.2	6.3

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SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6001	6002	6003	6004
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 504-K	Katalco 504-K	Katalco 504-K	Katalco 504-K
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	20.68	20.68	20.68
Temperatures, °C:				
Thermal Dissolver	450	450	460	440
Catalytic Reactor	425	425	425	425
Slurry Preheater	209	225	228	233
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	1.961	0.985	0.988	0.971
Raw Coal, kg/hr	0.529	0.266	0.267	0.262
Hydrogen, L/hr	702.	438.	438.	438.
SHSV, hr^{-1}; Thermal Dissolver	2.00	1.00	1.00	1.00
Catalytic Reactor	0.66	0.33	0.33	0.33
Hydrogen Treatrate, Wt. % Dry Coal	12.0	15.0	15.0	15.2
Catalyst Age (kg coal/kg catalyst)	28	34	39	45
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	53.8	73.8	73.8	68.3
Toluene Solubles plus Gases	76.2	87.6	88.0	84.9
THF Solubles plus Gases	88.7	92.2	92.0	91.3
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	2.9	4.6	5.7	3.6
C_2H_6	2.8	4.1	5.4	3.4
C_3H_8	1.9	2.9	4.5	2.3
C_4H_{10}	3.6	4.2	5.1	4.6
CO	0.2	0.2	0.2	0.1
H_2S	0.5	0.5	0.6	0.5
H_2O	3.8	5.6	6.5	5.0
NH_3	0.7	1.0	1.2	1.0
Oil	37.4	50.7	44.6	47.8
Asphaltenes	22.4	13.8	14.2	16.6
Preasphaltenes	12.5	4.6	4.0	6.4
Unreacted Coal	11.3	7.8	8.0	8.7
D. H_2 CONSUMPTION, WT. % OF maf COAL	5.0	7.3	7.3	7.2

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SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6005	6006	6007	6008
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 504-K	Katalco 504-K	Katalco 504-K	Katalco 504-K
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	17.24	20.68	17.24
Temperatures, °C:				
Thermal Dissolver	450	450	450	450
Catalytic Reactor	425	400	425	400
Slurry Preheater	232	195	192	230
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	0.993	1.462	1.466	0.986
Raw Coal, kg/hr	0.268	0.395	0.396	0.266
Hydrogen, L/hr	438.	660.	660.	438.
SHSV, hr^{-1}; Thermal Dissolver	1.00	1.50	1.50	1.00
Catalytic Reactor	0.33	0.50	0.50	0.33
Hydrogen Treatrate, Wt. % Dry Coal	14.9	15.2	15.1	15.0
Catalyst Age (kg coal/kg catalyst)	51	59	68	74
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	67.1	49.0	62.4	58.6
Toluene Solubles plus Gases	84.9	73.6	82.3	80.7
THF Solubles plus Gases	91.0	88.4	90.4	90.0
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	4.6	3.1	3.5	3.8
C_2H_6	4.1	2.8	2.9	3.4
C_3H_8	3.3	1.8	2.0	2.2
C_4H_{10}	6.0	4.2	4.3	5.4
CO	0.3	0.6	0.2	0.2
H_2S	0.5	0.5	0.5	0.5
H_2O	4.7	3.2	5.4	4.3
NH_3	1.0	0.6	0.8	0.8
Oil	42.6	32.2	42.8	38.0
Asphaltenes	17.8	24.6	19.9	22.1
Preasphaltenes	6.1	14.8	8.1	9.3
Unreacted Coal	9.0	11.6	9.6	10.0
D. H_2 CONSUMPTION, WT. % OF maf COAL	5.6	4.7	6.3	5.2

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6009	6010	6011	6012
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 504-K	Katalco 504-K	Katalco 504-K	Katalco 504-K
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	20.68	20.68	20.68
Temperatures, °C:				
Thermal Dissolver	450	450	450	450
Catalytic Reactor	425	400	425	400
Slurry Preheater	209	236	236	194
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	1.470	1.002	1.007	1.480
Raw Coal, kg/hr	0.397	0.271	0.272	0.400
Hydrogen, L/hr	660.	438.	444.	660.
SHSV, hr^{-1}; Thermal Dissolver	1.50	1.00	1.00	1.50
Catalytic Reactor	0.50	0.33	0.33	0.50
Hydrogen Treatrate, Wt. % Dry Coal	15.1	14.7	14.9	15.0
Catalyst Age (kg coal/kg catalyst)	83	90	97	106
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	59.8	61.2	68.9	50.5
Toluene Solubles plus Gases	81.2	81.9	85.3	74.5
THF Solubles plus Gases	90.0	90.5	91.2	88.6
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	3.8	3.7	4.4	2.9
C_2H_6	3.9	3.1	4.2	2.4
C_3H_8	2.3	1.8	3.0	1.8
C_4H_{10}	5.0	5.8	4.6	3.6
CO	0.2	0.2	0.1	0.2
H_2S	0.6	0.5	0.5	0.4
H_2O	5.1	4.7	5.6	3.8
NH_3	0.8	0.8	1.0	0.6
Oil	38.1	40.6	45.5	34.8
Asphaltenes	21.4	20.7	16.4	24.0
Preasphaltenes	8.8	8.6	5.9	14.1
Unreacted Coal	10.0	9.5	8.8	11.4
D. H_2 CONSUMPTION, WT. % OF maf COAL	5.3	5.6	6.7	5.1

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.		6014	6018	6019	6020
COAL TESTED		Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED		Katalco 504-K	Katalco 550	Katalco 550	Katalco 550
A.	OPERATING CONDITIONS				
	Unit back pressure, MPa	20.68	17.24	20.68	17.24
	Temperatures, °C:				
	Thermal Dissolver	450	450	450	450
	Catalytic Reactor	425	425	425	400
	Slurry Preheater	241	233	230	200
	Hot Separator	300	300	300	300
	Feedrates:				
	Slurry, kg/hr	0.984	0.989	1.078	1.517
	Raw Coal, kg/hr	0.266	0.267	0.291	0.410
	Hydrogen, L/hr	438.	438.	450.	660.
	SHSV, hr^{-1}; Thermal Dissolver	1.00	1.00	1.00	1.50
	Catalytic Reactor	0.33	0.33	0.33	0.50
	Hydrogen Treatrate, Wt. % Dry Coal	15.0	14.9	14.1	14.7
	Catalyst Age (kg coal/kg catalyst)	114	38	45	54
B.	CONVERSIONS OF maf COAL, WT. %				
	Pentane Solubles plus Gases	69.5	67.7	69.4	46.6
	Toluene Solubles plus Gases	85.9	85.0	85.3	71.3
	THF Solubles plus Gases	91.0	91.3	91.8	87.9
C.	PRODUCT YIELDS ON maf COAL, WT. %				
	CH_4	4.5	5.0	4.6	3.0
	C_2H_6	3.7	5.4	4.1	2.9
	C_3H_8	3.5	3.6	3.6	1.7
	C_4H_{10}	4.1	3.6	3.5	2.4
	CO	0.2	0.4	0.2	0.3
	H_2S	0.4	0.5	0.5	0.6
	H_2O	6.6	4.5	6.0	3.9
	NH_3	1.1	0.6	0.8	0.4
	Oil	45.4	44.1	46.1	31.4
	Asphaltenes	16.4	17.3	15.9	24.7
	Preasphaltenes	5.1	6.3	6.5	16.6
	Unreacted Coal	9.0	8.7	8.2	12.1
D.	H_2 CONSUMPTION, WT. % OF maf COAL	6.9	6.0	6.0	4.2

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6021	6022	6023	6024
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 550	Katalco 550	Katalco 550	Katalco 550
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	17.24	20.68	20.68
Temperatures, °C:				
Thermal Dissolver	450	450	450	450
Catalytic Reactor	425	400	400	425
Slurry Preheater	200	227	193	198
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	1.489	1.037	1.489	1.523
Raw Coal, kg/hr	0.402	0.280	0.402	0.411
Hydrogen, L/hr	660.	438.	660.	660.
SHSV, hr^{-1}; Thermal Dissolver	1.50	1.00	1.50	1.50
Catalytic Reactor	0.50	0.33	0.50	0.50
Hydrogen Treatrate, Wt. % Dry Coal	14.9	14.2	14.9	14.6
Catalyst Age (kg coal/kg catalyst)	67	73	81	91
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	56.7	53.3	48.7	57.1
Toluene Solubles plus Gases	78.7	78.1	73.6	79.4
THF Solubles plus Gases	89.0	89.6	88.8	89.4
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	3.7	3.6	2.8	3.5
C_2H_6	3.5	3.4	2.3	3.0
C_3H_8	2.7	2.5	1.9	2.4
C_4H_{10}	3.2	2.5	2.3	2.9
CO	0.3	0.3	0.2	0.2
H_2S	0.5	0.5	0.4	0.5
H_2O	4.6	4.5	4.1	5.3
NH_3	0.6	0.5	0.4	0.6
	37.6	35.5	34.3	38.7
Oil	22.0	24.8	24.9	22.3
Asphaltenes	10.3	11.5	15.2	10.0
Preasphaltenes	11.0	10.4	11.2	10.6
Unreacted Coal	5.4	4.2	5.2	5.7
D. H_2 CONSUMPTION, WT. % OF maf COAL				

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6025	6026	6027
COAL TESTED	Lingan HR-6	Lingan HR-6	Prince HB-D
CATALYST TESTED	Katalco 550	Katalco 550	Katalco 550
A. OPERATING CONDITIONS			
Unit back pressure, MPa	20.68	20.68	20.68
Temperatures, °C:			
Thermal Dissolver	450	450	440
Catalytic Reactor	400	425	425
Slurry Preheater	229	214	226
Hot Separator	300	300	300
Feedrates:			
Slurry, kg/hr	1.001	1.055	1.101
Raw Coal, kg/hr	0.270	0.285	0.297
Hydrogen, L/hr	438.	438.	468.
SHSV, hr^{-1}; Thermal Dissolver	1.00	1.00	1.12
Catalytic Reactor	0.33	0.33	0.37
Hydrogen Treatrate, Wt. % Dry Coal	14.7	14.0	14.6
Catalyst Age (kg coal/kg catalyst)	97	105	111
B. CONVERSIONS OF maf COAL, WT. %			
Pentane Solubles plus Gases	57.3	68.8	78.8
Toluene Solubles plus Gases	79.8	84.8	90.9
THF Solubles plus Gases	90.3	91.9	92.9
C. PRODUCT YIELDS ON maf COAL, WT. %			
CH_4	3.3	4.5	3.4
C_2H_6	2.9	3.9	3.4
C_3H_8	2.2	3.1	2.9
C_4H_{10}	2.7	3.1	3.1
CO	0.2	0.2	0.3
H_2S	0.4	0.5	2.0
H_2O	4.3	5.8	7.7
NH_3	0.5	0.7	0.7
Oil	40.8	47.0	55.3
Asphaltenes	22.5	16.0	12.1
Preasphaltenes	10.5	7.1	2.0
Unreacted Coal	9.7	8.1	7.1
D. H_2 CONSUMPTION, WT. % OF maf COAL	5.3	5.6	6.2

APPENDIX C

MATERIAL BALANCES FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

APPENDIX C

MATERIAL BALANCES FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6-CT-1	6-CT-2	6-CT-3
COAL TESTED	Lingan L	Lingan L	Lingan L
CATALYST TESTED	Katalco 504-K	Katalco 504-K	Katalco 504-K
MOISTURE, WT. %	1.29	1.31	1.33
ASH, WT. % DRY BASIS	4.27	4.27	4.24
HOURS ON STREAM	8.00	9.47	10.15
DURATION OF BALANCE PERIOD, HRS.	1.52	2.13	2.15
INPUT (g):			
H_2O	6.5	6.7	6.9
Ash	21.2	21.5	21.8
maf Coal	475.	482.	493.
Solvent	1508.	1530.	1565.
H_2	56.6	62.1	77.8
Total	2067.	2102.	2165.
OUTPUT (g):			
H_2O	32.3	40.1	41.1
Ash	15.0	14.7	16.9
Organic THF Insolubles[1]	34.3	31.9	31.9
THF Solubles[2]	1856.	1912.	1926.
H_2	28.7	32.1	46.7
Other Gases	70.4	94.5	88.9
Total	2047.	2125.	2152.
TOTAL MATERIAL RECOVERY, WT. %	99.0	101.1	99.4

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes

APPENDIX C

MATERIAL BALANCES FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6001	6002	6003	6004
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 504-K	Katalco 504-K	Katalco 504-K	Katalco 504-K
MOISTURE, WT. %	1.12	1.13	1.13	1.13
ASH, WT. % DRY BASIS	4.64	4.41	4.41	4.41
HOURS ON STREAM	9.50	9.50	9.50	9.57
DURATION OF BALANCE PERIOD, HRS.	1.18	2.02	1.97	2.00
INPUT (g):				
H_2O	7.0	6.1	5.9	5.9
Ash	28.8	23.4	22.9	22.9
maf Coal	591.	507.	496.	496.
Solvent	1694.	1451.	1419.	1418.
H_2	74.6	79.6	77.7	78.8
Total	2395.	2067.	2022.	2022.
OUTPUT (g):				
H_2O	29.5	34.7	38.0	30.9
Ash	25.4	15.8	17.5	16.2
Organic THF Insolubles[1]	66.7	39.7	39.8	43.3
THF Solubles[2]	2135.	1795.	1744.	1796.
H_2	45.0	42.6	41.5	43.2
Other Gases	74.6	89.4	112.6	76.7
Total	2376.	2017.	1992.	2006.
TOTAL MATERIAL RECOVERY, WT. %	99.2	97.6	98.6	99.2

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes

APPENDIX C

MATERIAL BALANCES FOR
TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6005	6006	6007	6008
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 504-K	Katalco 504-K	Katalco 504-K	Katalco 504-K
MOISTURE, WT. %	1.13	1.13	1.13	1.13
ASH, WT. % DRY BASIS	4.41	4.41	4.41	4.41
HOURS ON STREAM	9.50	9.50	9.50	9.62
DURATION OF BALANCE PERIOD, HRS.	2.02	1.35	1.33	2.00
INPUT (g):				
H_2O	6.1	6.0	6.0	6.0
Ash	23.6	23.2	23.0	23.2
maf Coal	512.	504.	499.	503.
Solvent	1464.	1441.	1426.	1439.
H_2	79.6	80.2	79.0	78.8
Total	2085.	2054.	2033.	2050.
OUTPUT (g):				
H_2O	30.4	22.2	33.1	27.8
Ash	16.5	19.0	18.4	16.9
Organic THF Insolubles[1]	46.2	58.5	48.0	50.4
THF Solubles[2]	1832.	1827.	1806.	1831.
H_2	50.9	56.5	47.8	52.5
Other Gases	101.2	68.2	70.9	82.1
Total	2077.	2051.	2024.	2061.
TOTAL MATERIAL RECOVERY, WT. %	99.6	99.9	99.6	100.5

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes

APPENDIX C

MATERIAL BALANCES FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6009	6010	6011	6012
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 504-K	Katalco 504-K	Katalco 504-K	Katalco 504-K
MOISTURE, WT. %	1.13	1.13	1.13	1.13
ASH, WT. % DRY BASIS	4.41	4.41	4.41	4.41
HOURS ON STREAM	10.10	9.60	9.50	9.63
DURATION OF BALANCE PERIOD, HRS.	1.12	2.00	2.03	1.32
INPUT (g):				
H_2O	5.0	6.1	6.2	6.0
Ash	19.4	23.6	24.1	23.0
maf Coal	420.	511.	522.	498.
Solvent	1202.	1463.	1493.	1426.
H_2	66.5	78.8	81.1	78.4
Total	1713.	2083.	2126.	2031.
OUTPUT (g):				
H_2O	26.4	30.0	35.6	24.9
Ash	15.2	17.7	17.1	19.4
Organic THF Insolubles[1]	42.0	48.5	46.0	56.6
THF Solubles[2]	1519.	1848.	1872.	1814.
H_2	44.2	50.4	45.9	53.1
Other Gases	69.5	81.3	94.1	58.9
Total	1716.	2076.	2111.	2027.
TOTAL MATERIAL RECOVERY, WT. %	100.2	99.7	99.3	99.8

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes

APPENDIX C

MATERIAL BALANCES FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6014	6018	6019	6020
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 504-K	Katalco 550	Katalco 550	Katalco 550
MOISTURE, WT. %	1.13	1.13	1.13	1.13
ASH, WT. % DRY BASIS	4.41	4.41	4.41	4.41
HOURS ON STREAM	9.50	8.35	9.00	8.50
DURATION OF BALANCE PERIOD, HRS.	2.00	1.35	2.03	1.35
INPUT (g):				
H_2O	6.0	4.1	6.7	6.2
Ash	23.2	15.7	25.8	24.1
maf Coal	502.	341.	559.	523.
Solvent	1437.	975.	1598.	1495.
H_2	78.8	53.2	82.2	80.2
Total	2047.	1389.	2272.	2129.
OUTPUT (g):				
H_2O	38.9	19.6	40.4	26.6
Ash	17.5	9.9	16.4	20.3
Organic THF Insolubles[1]	45.1	29.8	45.8	63.4
THF Solubles[2]	1809.	1198.	1971.	1914.
H_2	44.1	32.8	48.8	58.0
Other Gases	87.6	65.0	96.7	59.7
Total	2042.	1355.	2219.	2142.
TOTAL MATERIAL RECOVERY, WT. %	99.8	97.6	97.7	100.6

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes

APPENDIX C

MATERIAL BALANCES FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6021	6022	6023	6024
COAL TESTED	Lingan HR-6	Lingan HR-6	Lingan HR-6	Lingan HR-6
CATALYST TESTED	Katalco 550	Katalco 550	Katalco 550	Katalco 550
MOISTURE, WT. %	1.13	1.13	1.13	1.13
ASH, WT. % DRY BASIS	4.41	4.41	4.41	4.41
HOURS ON STREAM	12.00	8.20	8.22	9.67
DURATION OF BALANCE PERIOD, HRS.	1.52	2.02	1.33	1.33
INPUT (g):				
H_2O	6.9	6.4	6.0	6.2
Ash	26.6	24.7	23.3	23.8
maf Coal	577.	535.	505.	517
Solvent	1652.	1529.	1445.	1478
H_2	90.3	79.6	79.0	79.0
Total	2353.	2175.	2058.	2104.
OUTPUT (g):				
H_2O	33.3	30.4	26.6	33.8
Ash	24.9	19.5	19.7	20.4
Organic THF Insolubles[1]	63.7	55.4	56.4	54.6
THF Solubles[2]	2091.	1952.	1851.	1924.
H_2	59.2	56.9	52.9	49.3
Other Gases	83.3	71.5	52.4	67.3
Total	2355.	2186.	2059.	2149.
TOTAL MATERIAL RECOVERY, WT. %	100.1	100.5	100.0	102.2

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes

APPENDIX C

MATERIAL BALANCES FOR TWO-STAGE LIQUEFACTION EXPERIMENTS

RUN NO.	6025	6026	6027
COAL TESTED	Lingan HR-6	Lingan HR-6	Prince HB-D
CATALYST TESTED	Katalco 550	Katalco 550	Katalco 550
MOISTURE, WT. %	1.13	1.13	2.92
ASH, WT. % DRY BASIS	4.41	4.41	9.06
HOURS ON STREAM	8.40	10.10	8.05
DURATION OF BALANCE PERIOD, HRS.	1.87	2.08	2.02
INPUT (g):			
H_2O	5.7	6.7	17.5
Ash	22.0	25.8	52.8
maf Coal	478.	560.	530.
Solvent	1367.	1602.	1624.
H_2	73.7	82.0	85.1
Total	1946.	2277.	2309.
OUTPUT (g):			
H_2O	26.1	39.2	74.2
Ash	18.1	15.5	41.3
Organic THF Insolubles[1]	46.6	45.5	37.4
THF Solubles[2]	1726.	2009.	1988.
H_2	48.6	50.8	52.0
Other Gases	58.5	89.0	83.9
Total	1924.	2249.	2277.
TOTAL MATERIAL RECOVERY, WT. %	98.8	98.8	98.6

1 - Unreacted coal
2 - Oil, asphaltenes and preasphaltenes

APPENDIX D

RESULTS OF HPLC APPLIED TO COAL-DERIVED LIQUIDS

APPENDIX D

RESULTS OF HPLC APPLIED TO COAL-DERIVED LIQUIDS

During Phase VI, an open column method (ASTM D-2007) and a high performance liquid chromatography (HPLC) method were used for the separation of coal liquids into saturates, aromatics and polar compounds. Details pertaining to the HPLC method have been given in a previous report (6).

In the HPLC method, separation of coal liquids into "double bond" fractions was achieved. A number of model compounds were used to define the specific regions of the chromatograph as shown in Figure 5. The regions were defined by the number of double bonds in a given hydrocarbon molecule ranging from 0 (saturates) to 8 (tetracyclic PAH). Elution profiles for hydrogenated anthracene oil (9% H) and distillate samples from two-stage coal liquefaction experiments are shown in Figures 6 - 9.

Results obtained using both separation procedures are presented in Table 15. Excellent recovery was achieved in both methods. Agreement between the methods was fairly good. A comparison of HAO 61 with the distillates shows that the distillates have a higher concentration of saturates and a lower aromatics content. The results also illustrate that distillates from Run Nos. 6011, 6019, and 6027 contain higher concentrations of polar compounds than the reaction solvent used.

In future work, HPLC will be used to monitor hydrocarbon groups in start-up oil, recycle oil and distillate products.

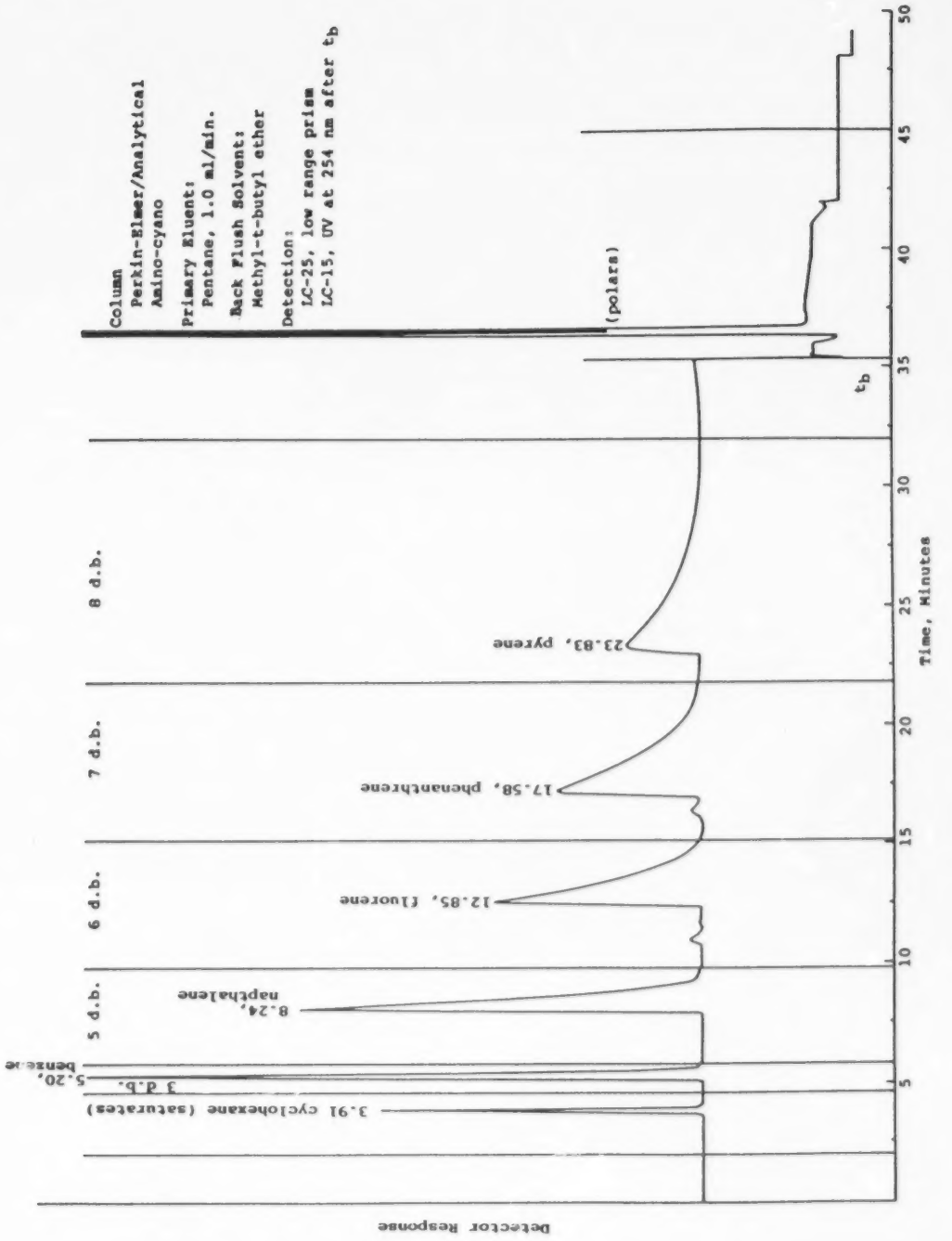

Figure 5 - HPLC ELUTION PROFILES FOR MODEL COMPOUNDS

Figure 6 - HPLC SEPARATION OF HYDROGENATED ANTHRACENE OIL WITH 9.03 WT.% HYDROGEN

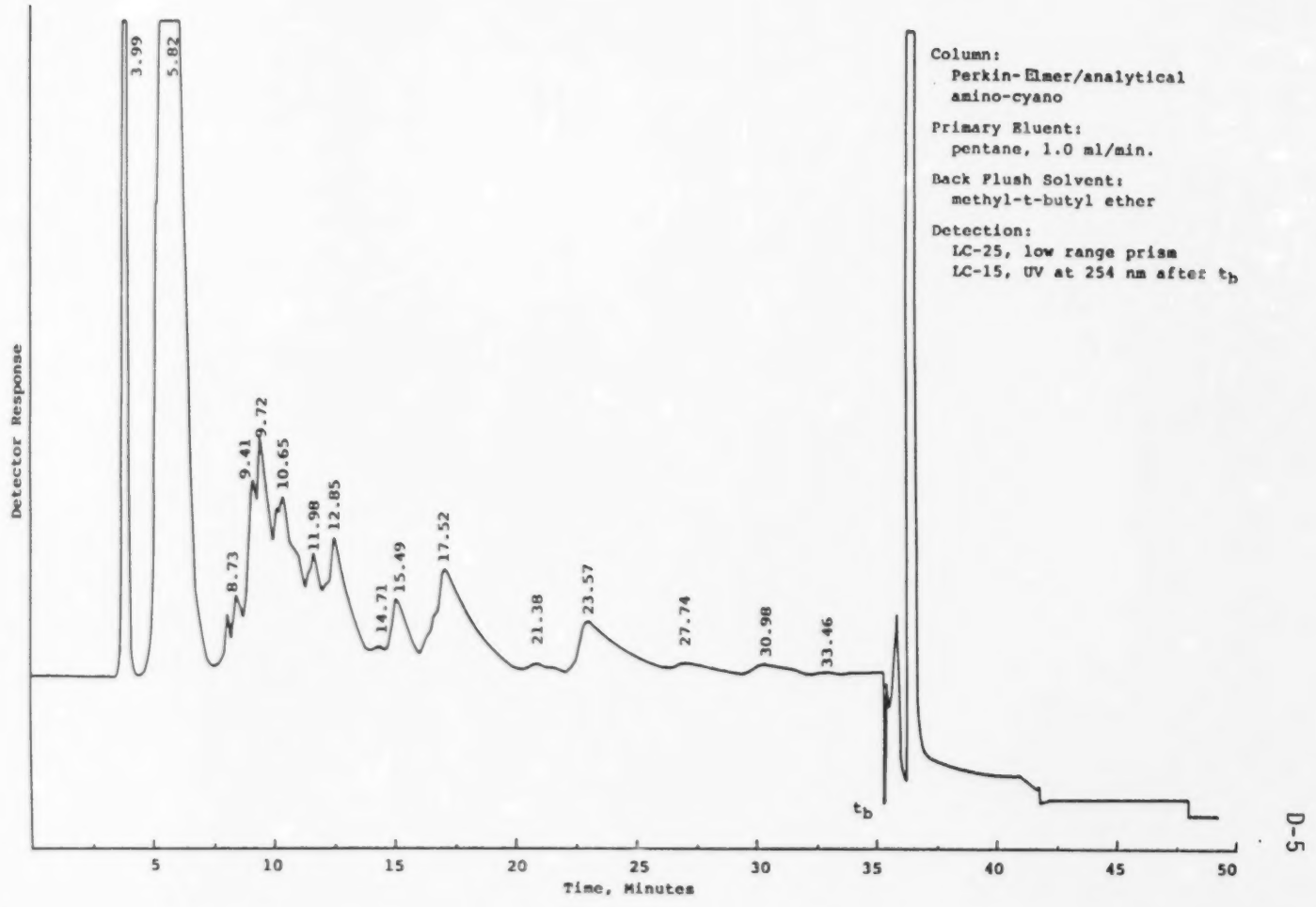

Figure 7 - HPLC SEPARATION OF DISTILLATE FROM RUN NO. 6011

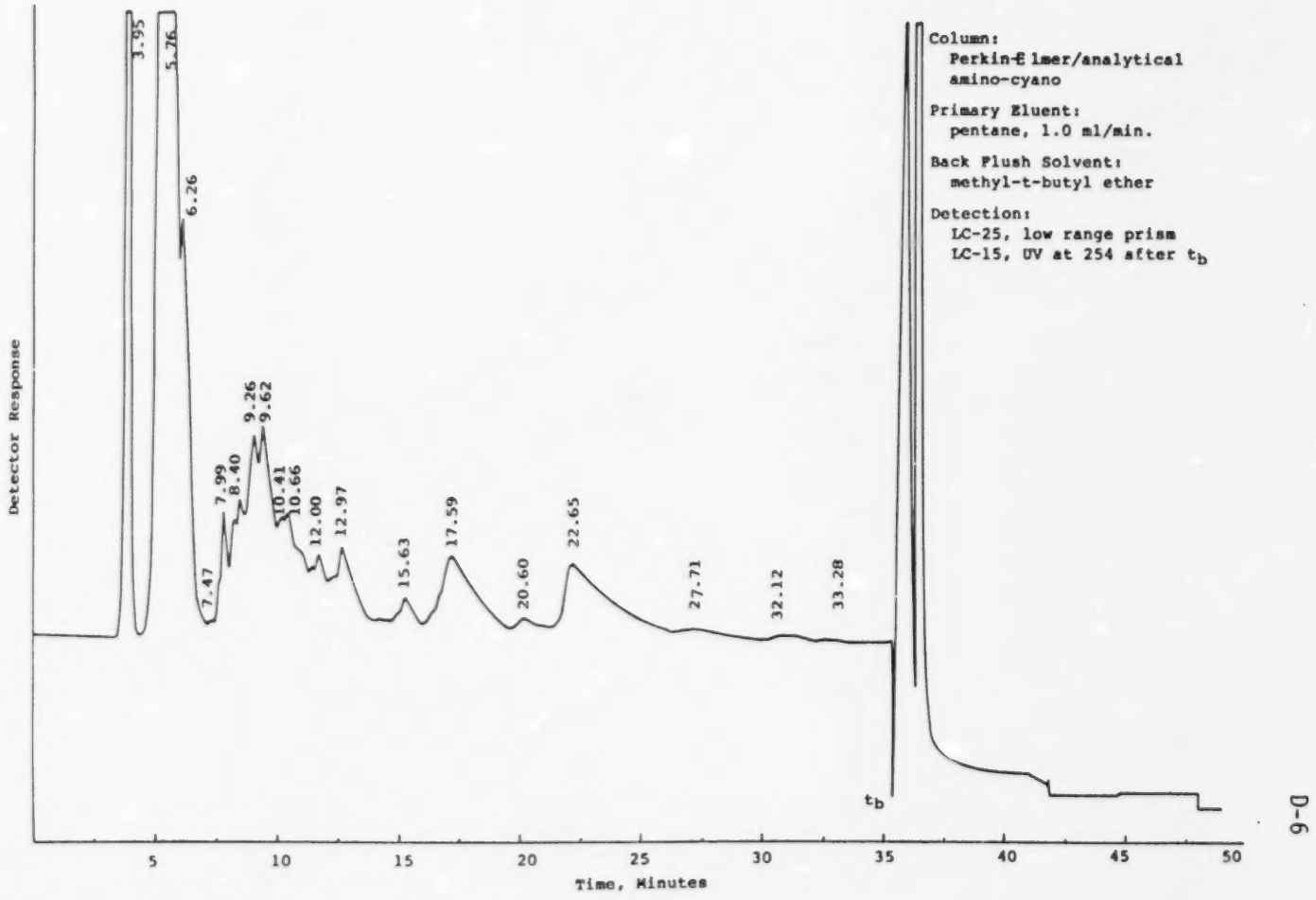

Figure 8 - HPLC SEPARATION OF DISTILLATE FROM RUN NO. 6019

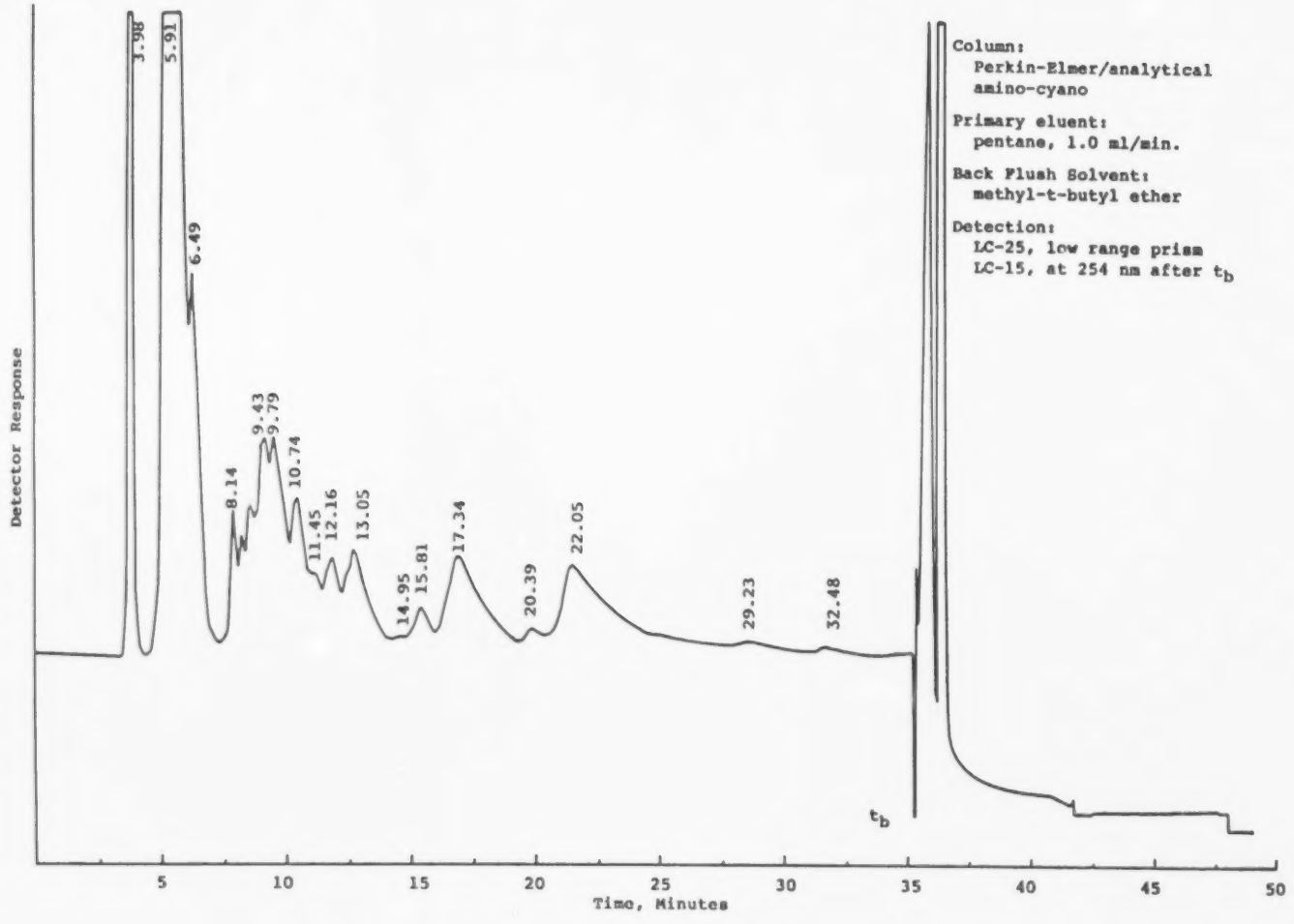

Figure 9 - HPLC SEPARATION OF DISTILLATE FROM RUN NO. 6027

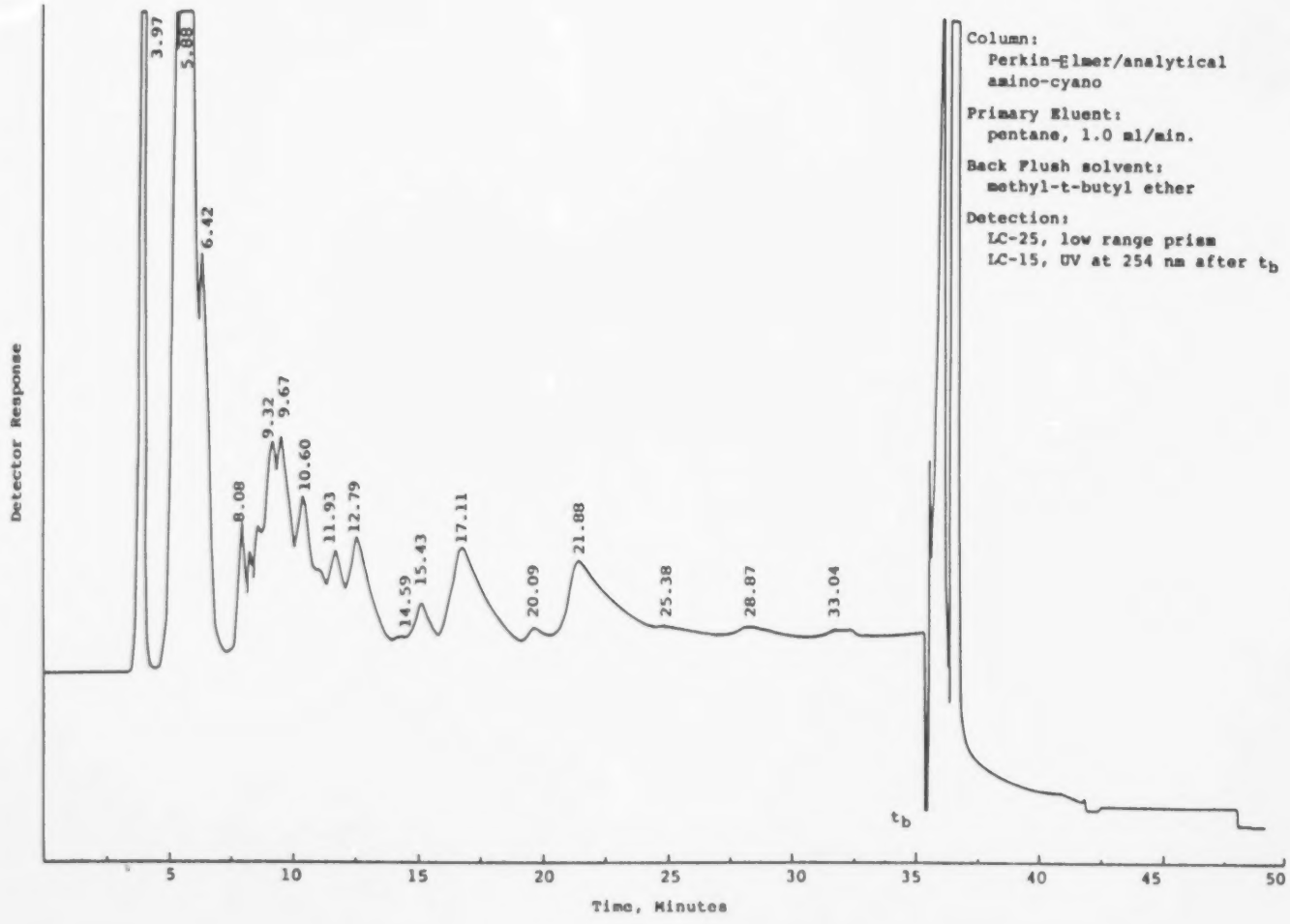

TABLE 15

CHROMATOGRAPHIC SEPARATION OF HYDROGENATED ANTHRACENE OIL AND DISTILLATE FRACTIONS FROM SELECTED COAL LIQUEFACTION EXPERIMENTS

SAMPLE DESCRIPTION	HAO 61		RUN NO. 6011 DISTILLATE		RUN NO. 6019 DISTILLATE		RUN NO. 6027 DISTILLATE	
SEPARATION METHOD	HPLC	ASTM D-2007	HPLC	ASTM D-2007	HPLC	ASTM D-2007	HPLC	ASTM D-2007
Saturates, Wt. %	13.93	11.14	19.53	16.12	15.75	13.99	13.60	13.00
Aromatics, Wt. %	82.05	84.97	75.25	76.22	77.26	80.70	81.00	81.37
3 double bonds	10.17		10.36		11.04		11.33	
5 double bonds	28.14		30.77		29.10		31.00	
6 double bonds	21.36		15.72		15.72		14.67	
7 double bonds	10.85		9.70		10.70		10.67	
8 double bonds	11.53		8.70		10.70		13.33	
Polars, Wt. %	3.39	2.03	6.02	3.99	7.02	4.56	6.33	5.19
Recovery, Wt. %	99.37	98.14	100.80	96.33	100.03	99.25	100.93	99.56

APPENDIX E

STATISTICAL ANALYSIS OF 2^4 FACTORIAL EXPERIMENT ON LINGAN HARBOUR SEAM COAL

APPENDIX E

RESPONSE FACTOR: GASES

SLURRY FEEDRATE (kg/hr)	400°C				425°C			
	17.24 MPa		20.68 MPa		17.24 MPa		20.68 MPa	
	C_1	C_2	C_1	C_2	C_1	C_2	C_1	C_2
1	20.6	17.8	20.6	16.5	24.5	23.6	23.4	23.3
1.5	16.8	15.2	15.7	14.4	21.7	19.1	19.6	18.4

$\Sigma x = 311.2$ $\quad C = \frac{(\Sigma x)^2}{n} = \frac{(311.2)^2}{16} = 6052.8400$

$SS_{Total} = \Sigma x^2 - C = 158.1800$

$SS_{Temperature} = 49{,}070.72/8 - C = 81.0000$

$SS_{Pressure} = 48{,}450.10/8 - C = 3.4225$

$SS_{Feedrate} = 48{,}854.90/8 - C = 54.0225$

$SS_{Catalyst} = 48{,}529.30/8 - C = 13.3225$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	81.0000	81.0000	138.9365	.99	+ 36.0
Pressure	1	3.4225	3.4225	5.8705	.95	- 7.4
Feedrate	1	54.0225	54.0225	92.6630	.99	- 29.4
Catalyst	1	13.3225	13.3225	22.8516	.99	- 14.6
Residual	11	6.4125	0.5830			
Total	15	158.1800				

* n = 16

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 311.2$

$\bar{x} = \frac{\Sigma x}{n} = \frac{311.2}{16} = 19.5$

$S^2 = \frac{SS}{df} = \frac{6.4125}{11} = 0.5830$

$S = \sqrt{0.5830} = 0.76$

$S_{\bar{x}} = \frac{S}{\sqrt{n}} = \frac{0.76}{\sqrt{16}} = 0.2$

$\therefore$ the mean of 16 results at the 2σ confidence interval is:

19.5 ± 0.4

APPENDIX E

RESPONSE FACTOR: OIL

SLURRY FEEDRATE (kg/hr)	400°C				425°C			
	17.24 MPa		20.68 MPa		17.24 MPa		20.68 MPa	
	C_1	C_2	C_1	C_2	C_1	C_2	C_1	C_2
1	38.0	35.5	40.6	40.8	42.6	44.1	45.5	46.1
1.5	32.2	31.4	34.8	34.3	38.1	37.6	42.8	38.7

$\Sigma x = 623.1$ $\quad C = \frac{(\Sigma x)^2}{n} = \frac{(623.1)^2}{16} = 24{,}265.8506$

$SS_{Total} = \Sigma x^2 - C = 311.6594$

$SS_{Temperature} = 195{,}274.01/8 - C = 143.4007$

$SS_{Pressure} = 194{,}417.21/8 - C = 36.3007$

$SS_{Feedrate} = 195{,}064.25/8 - C = 117.1807$

$SS_{Catalyst} = 194{,}145.41/8 - C = 2.3257$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	143.4007	143.4007	126.6791	.99	+ 47.9
Pressure	1	36.3007	36.3007	32.0678	.99	+ 24.1
Feedrate	1	117.1807	117.1807	103.5165	.99	- 43.3
Catalyst	1	2.3257	2.3257	2.0545	.75	- 6.1
Residual	11	12.4516	1.1320			
Total	15	311.6594				

* n = 16

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 623.1$

$x = \frac{\Sigma x}{n} = \frac{623.1}{16} = 38.9$

$S^2 = \frac{SS}{df} = \frac{12.4516}{11} = 1.1320$

$S = \sqrt{1.1320} = 1.06$

$S_x = \frac{S}{\sqrt{n}} = \frac{1.06}{\sqrt{16}} = 0.3$

$\therefore$ the mean of 16 results at the 2σ confidence interval is:

38.9 ± 0.6

APPENDIX E

RESPONSE FACTOR: ASPHALTENES

SLURRY FEEDRATE (kg/hr)	400°C				425°C			
	17.24 MPa		20.68 MPa		17.24 MPa		20.68 MPa	
	C_1	C_2	C_1	C_2	C_1	C_2	C_1	C_2
1	22.1	24.8	20.7	22.5	17.8	17.3	16.4	15.9
1.5	24.6	24.7	24.0	24.9	21.4	22.0	19.9	22.3

$\Sigma x = 341.3$ $\quad C = \frac{(\Sigma x)^2}{n} = \frac{(341.3)^2}{16} = 7280.3556$

$SS_{Total} = \Sigma x^2 - C = 142.2544$

$SS_{Temperature} = 58{,}865.89/8 - C = 77.8807$

$SS_{Pressure} = 58{,}275.65/8 - C = 4.1007$

$SS_{Feedrate} = 58{,}588.69/8 - C = 43.2307$

$SS_{Catalyst} = 58{,}270.97/8 - C = 3.5157$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	77.8807	77.8807	63.3331	.99	- 35.3
Pressure	1	4.1007	4.1007	3.3347	.90	- 8.1
Feedrate	1	43.2307	43.2307	35.1555	.99	+ 26.3
Catalyst	1	3.5157	3.5157	2.8590	.75	+ 7.5
Residual	11	13.5266	1.2297			
Total	15	142.2544				

* n = 16

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 341.3$

$x = \frac{\Sigma x}{n} = \frac{341.3}{16} = 21.3$

$S^2 = \frac{SS}{df} = \frac{13.5266}{11} = 1.2297$

$S = \sqrt{1.2297} = 1.11$

$S_x = \frac{S}{\sqrt{n}} = \frac{1.11}{\sqrt{16}} = 0.3$

∴ the mean of 16 results at the 2σ confidence interval is:

21.3 ± 0.6

APPENDIX E

RESPONSE FACTOR: PREASPHALTENES

SLURRY FEEDRATE (kg/hr)	400°C				425°C			
	17.24 MPa		20.68 MPa		17.24 MPa		20.68 MPa	
	C_1	C_2	C_1	C_2	C_1	C_2	C_1	C_2
1	9.3	11.5	8.6	10.5	6.1	6.3	5.9	6.5
1.5	14.8	16.6	14.1	15.2	8.8	10.3	8.1	10.0

$\Sigma x = 162.6$ $\quad C = \frac{(\Sigma x)^2}{n} = \frac{(162.6)^2}{16} = 1652.4225$

$SS_{Total} = \Sigma x^2 - C = 178.0775$

$SS_{Temperature} = 13,964.36/8 - C = 93.1225$

$SS_{Pressure} = 13,230.90/8 - C = 1.4400$

$SS_{Feedrate} = 13,770.50/8 - C = 68.8900$

$SS_{Catalyst} = 13,282.10/8 - C = 7.8400$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	93.1225	93.1225	150.9768	.99	- 38.6
Pressure	1	1.4400	1.4400	2.3346	.75	- 4.8
Feedrate	1	68.8900	68.8900	111.6894	.99	+ 33.2
Catalyst	1	7.8400	7.8400	12.7108	.99	+ 11.2
Residual	11	6.7850	0.6168			
Total	15	178.0775				

* n = 16

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 162.6$

$\bar{x} = \frac{\Sigma x}{n} = \frac{162.6}{16} = 10.2$

$S^2 = \frac{SS}{df} = \frac{6.7850}{11} = 0.6168$

$S = \sqrt{0.6168} = 0.79$

$S_{\bar{x}} = \frac{S}{\sqrt{n}} = \frac{0.79}{\sqrt{16}} = 0.2$

∴ the mean of 16 results at the 2σ confidence interval is:

10.2 ± 0.4

APPENDIX E

RESPONSE FACTOR: H_2 CONSUMPTION

SLURRY FEEDRATE (kg/hr)	400°C				425°C			
	17.24 MPa		20.68 MPa		17.24 MPa		20.68 MPa	
	C_1	C_2	C_1	C_2	C_1	C_2	C_1	C_2
1	5.2	4.2	5.6	5.3	5.6	6.0	6.7	6.0
1.5	4.7	4.2	5.1	5.2	5.3	5.4	6.3	5.7

$\Sigma x = 86.5$ $\quad C = \frac{(\Sigma x)^2}{n} = \frac{(86.5)^2}{16} = 467.6406$

$SS_{Total} = \Sigma x^2 - C = 6.9494$

$SS_{Temperature} = 3{,}769.25/8 - C = 3.5157$

$SS_{Pressure} = 3{,}755.17/8 - C = 1.7557$

$SS_{Feedrate} = 3{,}744.77/8 - C = 0.4557$

$SS_{Catalyst} = 3{,}744.25/8 - C = 0.3907$

ANALYSIS OF VARIANCE

SOURCE	df	SS	MS	F	CONFIDENCE LEVEL	EFFECT*
Temperature	1	3.5157	3.5157	46.5040	.99	+ 7.5
Pressure	1	1.7557	1.7557	23.2235	.99	+ 5.3
Feedrate	1	0.4557	0.4557	6.0278	.95	- 2.7
Catalyst	1	0.3907	0.3907	5.1680	.95	- 2.5
Residual	11	0.8316	0.0756			
Total	15	6.9494				

* n = 16

CALCULATION OF CONFIDENCE INTERVAL

$\Sigma x = 86.5$

$\bar{x} = \frac{\Sigma x}{n} = \frac{86.5}{16} = 5.4$

$S^2 = \frac{SS}{df} = \frac{0.8316}{11} = 0.0756$

$S = \sqrt{0.0756} = 0.27$

$S_{\bar{x}} = \frac{S}{\sqrt{n}} = \frac{0.27}{\sqrt{16}} = 0.1$

∴ the mean of 16 results at the 2σ confidence interval is:

5.4 ± 0.2

APPENDIX F

OPERATING PROCEDURE FOR

TWO-STAGE CONTINUOUS COAL LIQUEFACTION UNIT

This procedure is normally carried out the day before a run.

1. Open valves 1, 7, 11, 14, 15, 116, 119 and 122.

2. Depressure the system to 345 kPa by opening valves 17 and 18. Close valves 17 and 18.

3. Open valve 2 and valve 8 and pressurize the system as well as the 7.6 liter emergency blowdown vessel to 4.14 MPa. Close valve 8 and continue pressurizing the system until the pressure is at the desired level either 10.34 MPa or 13.79 MPa. Close valve 2.

4. Leave for a few hours and monitor pressure.

5. Set the forward pressure regulator and the back pressure regulator to the desired settings.

6. Turn on the cooling water to the magnedrive stirrers.

7. Set the controllers at the desired settings for both the first-stage and second-stage reactors.

	Stage 1	Stage 2
reactor controller °C	343	288
furnace controller °C	538	450
zone controllers, %	60	100

8. Set the automatic timer to activate the reactor furnace temperature controllers according to temperature programming schedule. Turn the reactor temperature controller switches to the "ON" position and ensure they are not activated.

9. Set the temperature controller for high pressure separator No. 1, V-5, at 300 °C.

10. Set the Automatic timer to activate V-5 temperature controller at 4 hours prior to anticipated start-up. Turn the controller switch to the "ON" position and ensure it is not activated.

11. Ensure that alarm switch No. 14 is deactivated.

Start-Up

1. Gradually increase the furnace controllers for stage 1 and stage 2 to the desired levels. NEVER allow the heating rate of the reactors to exceed 120°C per hour.

2. Turn on heating tapes on slurry transfer lines:

   No. 1 - 33 %, at start of run gradually increase to 100 %

   No. 2 - 33%, at start of run gradually increase to 66 %

   No. 3 - 33 %

   No. 4 - 33 %

3. Activate heater on high pressure separator No. 2, V-7.

4. Turn on condenser cooling water.

5. Activate K-Ray electronics for liquid level measurement system.

6. After one hour open the window on the radioactive source.

7. Turn on the hydrogen flow control system fifteen minutes prior to run.

   Procedure

   a) Turn on the power.
   b) Open valve 3.
   c) Set the air pressure at 138 kPa.
   d) Adjust the setpoint knob until the ball is in the center, then switch from manual to automatic.
   e) Set the pen to the desired value.
   f) Open valve 123 and close valve 122.
   g) Allow the hydrogen flow control system to stabilize.
   h) Monitor exit gas flow rate for 15 minutes.

8. Purge the feed tank with nitrogen 15 minutes prior to slurry addition. Maintain a small nitrogen flow throughout run.

9. Charge V-1 with the slurry. Open valve 101 and turn on the circulating pump.

10. Turn off the circulating pump for 15 seconds and then turn circulating pump back on. This fills void spaces or gas pockets in the pump with slurry and ensures proper weight readings on the toledo scale. Turn on the alarm switch No. 14 in the side panel.

11. When the temperatures of the reactors are at the desired levels open valve 102 and purge the high pressure metering pump with slurry until the liquid is gas free.

Run

12. Open the plant valve (valve 6) and activate the high pressure metering pump.

13. Turn on the V-3, V-4 and V-5 magnedrive stirrers.

14. Turn on the cooling fan for magnedrive stirrer on V-4.

15. Monitor the pressures, temperatures, slurry feedrate and vent gas flow rates throughout the run.

16. A rapid increase in the reactor 1 temperature will occur 10 to 20 minutes after the high pressure metering pump has been activated. When the reactor temperature reaches 4°C below the target temperature turn the furnace temperature controller down until the red light goes off. Increase the furnace temperature controller back to the original setpoint when reactor 1 temperature levels off.

17. Open valves 111, 107, 104 and 105 and purge V-9 with nitrogen, 10 minutes prior to step 18.

18. After 2.3 kg of slurry has been pumped, open valve 22.

19. Set timer K-5 for the automatic letdown (ALD) system to maintain a constant level in V-5.

20. Open valve 19.

21. Set the selector switch on "Automatic During High Alert" and activate the automatic letdown system.

22. After the first ALD cycle (mark this on data sheet) close valve 105.

23. Sample vent gas streams 15 minutes prior to steady state sampling period.

24. Record the toledo scale weight and deactivate the circulating pump for 15 seconds 15 minutes prior to steady state sampling period. Reactivate the circulating pump and record the toledo scale weight.

25. Drain liquid from the separator overheads knock-out (V-7) periodically and 5 minutes prior to steady state sampling period by opening and closing valves 17 and 18 sequentially.

26. To initiate the steady state sampling period, record the toledo scale weight after valve "A" has closed on the final ALD cycle of the slop period.

27. Record the wet test meter reading and the circulating pump pan weight.

28. Open valves 108 and 23. Close valves 22 and 107. Open valve 106 and start a nitrogen purge.

29. On the next ALD cycle, the first in the steady state sampling period, mark the stroke made on the hydrogen flow recorder.

30. Close valve 106.

31. Open valve 120 and drain V-15. Close valve 120.

32. Sample vent gas streams 15 minutes prior to end of steady state sampling period.

33. Five minutes prior to end of run, drain liquid from separator overheads knock-out, V-7, by opening and closing valves 17 and 18 sequentially.

34. At the end of the period, record the toledo scale weight after valve "A" has closed on the final ALD cycle of the sampling period. Close valve 19 and deactivate the automatic letdown system.

35. Record the wet test meter reading and the circulating pump pan weight. Empty contents of V-15.

36. Turn the reactor controller setpoints on stage 1 and stage 2 reactors down to 260°C. Turn the furnace controller setpoints on stage 1 and stage 2 reactors down to 204°C. Reduce voltage to heating tapes to 33% on slurry transfer lines. Turn off high pressure separator heaters.

37. Record the toledo scale weight and deactivate the circulating pump for 15 seconds. Reactivate the circulating pump and record the toledo scale weight.

38. Shut off the hydrogen flow control system.

Procedure

a) Turn the regulator knob until the ball is in the center.
b) Switch from automatic to manual.
c) Shut off the air flow.
d) Close valve 3.
e) Turn off the power.

39. Deactivate the high pressure metering pump.

40. Close the plant valve (valve No. 6) and the interstage valve (valve No. 7) and valves 11 and 14.

41. Transfer contents of V-3 into the 7.6 liter high pressure vessel (which has been pressurized to 4.14 MPa hydrogen) through valve 8. Leave valve 8 open for 5 minutes and then close valve 8.

42. Transfer contents of V-4 into the 7.6 liter pressure vessel through valve 9. Leave valve 9 open for 5 minutes and then close valve 9.

43. Open valve 109. Manually withdraw slurry product from V-5 by opening valves 25 and then 24. Close valves 24 and then 25. Close valve 109. Open valve 13 and repressure V-5 to 13.79 MPa and then repeat step 43.

44. Open valve 120 and drain contents of V-15. Close valve 120.

45. Deactivate the circulating pump and drain slurry from the feed tank.

46. Open valves 14 and 15 to equalize the pressure.

47. Note: DO NOT LET THE PRESSURE IN THE CATALYTIC REACTOR FALL BELOW 6.89 MPa. IF IT FALLS TO 6.89 MPa, SLOWLY OPEN VALVE 11 UNTIL THE PRESSURE IN STAGE 2 REACHES 8.27 MPa. CLOSE VALVE 11.

48. Note: DO NOT LET THE TEMPERATURE IN STAGE 2 FALL BELOW 288°C.

49. Open valve 2 and add high pressure hydrogen to V-3 (stage 1 reactor) until its pressure is 689 kPa in excess of the stage 2 reactor pressure. Close valve 2.

50. Open the interstage valve (valve 7) to equalize system pressure.

51. Open valve 2 and add high pressure hydrogen to stage 1 and 2 reactors until the pressure is 689 kPa in excess of the down stream pressure. Close valve 2.

52. Open valve 11 to equalize the pressure throughout the system.

53. Open valve 122 and close valve 123.

Flush Cycle and Shut-Down

54. Reactivate the reactor controller setpoints in both reactors to 316°C. Reactivate the furnace controller setpoints in both reactors to 427°C. Turn on the heater on separator No. 1 to 300°C . Increase voltage on line heaters to 66 % for the preheater and 33% for the interstage and slurry transfer lines.

55. Close valve 101 and 102 and add flush oil to V-1, open valve 101 and turn on the circulating pump.

56. Open valve 102 and purge the high pressure metering pump with oil until the liquid is gas free. Open the plant valve and activate the high pressure metering pump.

57. Note: Keep temperatures of stage 1 and stage 2 reactors between 288°C and 316°C during flush cycle.

58. Open valve 111 and 120 and drain V-15. Close valve 111.

59. When approximately 3.0 kg of oil has been pumped into V-5, open valves 111, 108, 23 and 19. Activate the ALD system allowing it to operate until at least 6 kg of flush oil has been pumped. Close valve 19, allow one more cycle. Deactivate the ALD system and close valves 23, 108 and 111.

60. Pull plug out for electronics on ALD system and close window on radioactive source.

61. Deactivate the metering pump.

62. Shut off all heating tapes, heaters and the reactor furnaces.

63. Turn off the condenser cooling water. Leave magnedrive water on overnight.

64. Close the plant valve, the interstage valve and valves 11 and 14.

65. Transfer contents of V-3 into the 7.6 liter high pressure vessel by opening valve 8. Let drain for 5 minutes. Prior to closing valve 8, open valve 2 and slowly add pressure to V-3 and the 7.6 liter vessel until the pressure is 4.83 MPa, then close valve 2 and valve 8.

66. Transfer contents of V-4 into 7.6 liter high pressure vessel by opening valve 9. Let stand for 5 minutes then close valve 9.

67. Withdraw oil from V-5.

68. Follow steps 49, 50, 51 and 52. Then open valve 2 and add hydrogen to the system until the pressure is 13.79 MPa.

70. Drain flush oil from V-1.

71. Turn off electronics for toledo scale and digital pressure meter.

72. Shut off alarm switch No. 14 in side panel.

POST-RUN OPERATIONS

This procedure is normally carried out the day after the run.

1. Close the interstage valve (valve No. 7) and valves 11 and 14.

2. Depressure the 7.6 liter pressure vessel to 4.83 MPa.

3. Empty contents of V-3 by opening valve 8. Close valve 8.

4. Depressure the 7.6 liter pressure vessel to 4.83 MPa.

5. Empty contents of V-4 by opening valve 9. Close valve 9.

6. Drain contents of V-5 by opening valves 110, 26 and 24. Close valves 24, 26 and 110.

7. Follow steps 49 to 52 in the ACTUAL OPERATION.

8. Depressure the 7.6 liter pressure vessel to 1.38 MPa. Empty contents of the 7.6 liter vessel into the 10 liter low pressure vessel.

9. Follow steps 1 to 4 on page 1.